

Effect of Mn^{2+} Substitution on the Structural, Magnetic, and Electrical Properties of Nanocrystalline $\text{Mg}_{0.2}\text{Mn}_{\square}\text{Co}_{0.8-\square}\text{Fe}_2\text{O}_4$ Ferrites

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Effect of Mn²⁺ Substitution on the Structural, Magnetic, and Electrical Properties of Nanocrystalline Mg_{0.2}Mn_xCo_{0.8-x}Fe₂O₄ Ferrites

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Abstract

Nanocrystalline Mg_{0.2}Mn_xCo_{0.8-x}Fe₂O₄ ferrites with Mn concentrations of $x = 0.0, 0.2, 0.4, 0.6, \text{ and } 0.8$ were synthesized by the citrate-assisted sol–gel auto-combustion method to systematically investigate the influence of Mn²⁺ substitution on their structural, morphological, magnetic, and electrical properties. X-ray diffraction (XRD) patterns together with Rietveld refinement confirmed the formation of a cubic spinel ferrite structure without secondary phases. The structural parameters exhibited a composition-dependent evolution with Mn incorporation. The crystallite size decreased from approximately 50 to 20 nm with increasing Mn content, indicating progressive crystallite refinement, while the lattice parameter and hopping lengths showed an overall increasing tendency associated with the substitution of Co²⁺ by the comparatively larger Mn²⁺ ions and the accompanying redistribution of cations between tetrahedral and octahedral sites. Williamson–Hall analysis revealed composition-dependent microstrain, demonstrating the influence of Mn substitution on lattice distortion and defect-related structural characteristics. The calculated structural parameters, including X-ray density, dislocation density, and unit-cell volume, further established the correlation between Mn concentration and the evolution of the spinel lattice. FESEM and AFM investigations revealed nanoscale particles with composition-dependent agglomeration, surface morphology, and topographical characteristics. FTIR spectra exhibited the characteristic metal–oxygen vibrational bands associated with the tetrahedral and octahedral sites of the spinel lattice,

confirming the structural framework of the synthesized ferrites. Magnetic measurements performed at 300 and 5 K demonstrated ferrimagnetic behavior for the investigated compositions. The M–H hysteresis loops showed relatively narrow loops at room temperature, whereas a pronounced increase in coercivity and remanent magnetization was observed at low temperature, reflecting enhanced magnetocrystalline anisotropy and reduced thermal fluctuations. The corresponding dM/dH curves provided further evidence of the temperature-dependent magnetic reversal mechanism. ZFC–FC magnetization measurements at different applied fields revealed significant thermomagnetic irreversibility, with stronger separation between the ZFC and FC branches at lower magnetic fields, indicating the contribution of magnetic interactions, anisotropy-energy barriers, and thermally activated spin dynamics. Impedance spectroscopy demonstrated strong frequency- and temperature-dependent electrical behavior characteristic of semiconducting ferrites. The decrease in impedance with increasing frequency and temperature suggests thermally activated charge-carrier hopping, while the broad and frequency-dependent relaxation features indicate non-Debye-type dielectric relaxation arising from the heterogeneous grain–grain-boundary structure. Among the investigated compositions, $x = 0.6$ exhibited particularly pronounced impedance and relaxation characteristics, suggesting enhanced grain-boundary polarization and charge-carrier localization. Overall, the results establish clear structure–morphology–magnetism–electrical property correlations induced by Mn^{2+} substitution and demonstrate that controlled compositional modification of Mg–Co ferrites provides an effective route for tailoring their magnetic anisotropy, magnetic reversal, charge transport, and relaxation behavior. The obtained multifunctional characteristics make $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites attractive for potential applications in magnetic sensors, high-frequency devices, electromagnetic components, and related multifunctional magnetic technologies..

Keywords: XRD, Magnetitic, Ferrite, Sol- Gel Method, Raman

1. Introduction

Spinel ferrites with the general formula MFe_2O_4 (where M represents divalent metal ions) have attracted considerable attention owing to their remarkable structural stability, magnetic behavior, electrical characteristics, and chemical durability[1], [2], [3], [4], [5], [6]. These materials exhibit a cubic spinel structure in which metal cations occupy tetrahedral (A) and octahedral (B) interstitial sites within a close-packed oxygen lattice. The distribution of cations among these sites strongly influences the structural, magnetic, optical, and electrical properties of ferrites, making them suitable for a wide range of technological applications including high-frequency devices, magnetic recording media, sensors, microwave absorbers, catalysis, biomedical systems, and electromagnetic interference shielding[7], [8], [9], [10], [11], [12], [13], [14], [15].

Among the various spinel ferrites, cobalt ferrite ($CoFe_2O_4$) is recognized for its high magnetocrystalline anisotropy, moderate saturation magnetization, excellent chemical stability, and large coercivity[16], [17], [18], [19], [20]. These properties make cobalt ferrite a promising material for permanent magnets and high-density magnetic storage applications. However, its relatively high coercivity and electrical conductivity may limit its performance in certain applications. Consequently, cation substitution has been extensively employed as an effective strategy to tailor the physical properties of cobalt ferrite[21], [22], [23], [24], [25]. The incorporation of suitable metal ions can modify the cation distribution, super-exchange interactions, grain growth behavior, and electronic structure, thereby enhancing the desired characteristics[26], [27], [28], [29].

Magnesium ferrite ($MgFe_2O_4$), a partially inverse spinel ferrite, exhibits low dielectric losses, good chemical stability, and high electrical resistivity. The introduction of Mg^{2+} ions into cobalt ferrite can reduce eddy current losses and improve the material's suitability for high-

frequency applications. On the other hand, manganese ferrite (MnFe_2O_4) possesses relatively high saturation magnetization and soft magnetic behavior due to the strong magnetic moment of Mn^{2+} ions. Substituting Mn^{2+} ions into cobalt-based ferrites can significantly influence the A–B and B–B super-exchange interactions, leading to modifications in magnetic ordering, anisotropy, and electrical transport properties[30], [31], [32].

Recently, nanocrystalline ferrites have gained increasing interest because their reduced particle size and large surface-to-volume ratio introduce unique physical phenomena that are absent in bulk materials. At the nanoscale, factors such as surface spin disorder, finite-size effects, and altered cation distribution can substantially affect the magnetic and optical responses. Therefore, understanding the influence of controlled cation substitution in nanocrystalline ferrites is essential for optimizing their performance in advanced technological applications[27], [33], [34], [35], [36], [37].

In the present work, a series of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ (Where $x=0.0, 0.2, 0.4, 0.6$, and 0.8) nanocrystalline ferrites were synthesized using the sol–gel auto-combustion method. The concentration of Mg^{2+} ions was kept constant while Mn^{2+} ions progressively replaced Co^{2+} ions in the spinel lattice. The effect of Mn substitution on the structural, morphological, optical, electrical, and magnetic properties of the prepared ferrites was systematically investigated. The correlation between cation substitution, crystallographic parameters, microstructure, and functional properties was analyzed to evaluate the suitability of these materials for magnetic and electronic device applications.

2. Experimental

2.1. Synthesis of Ferrite Nanoparticles by Sol–Gel Method

The nanocrystalline ferrites of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ (where $x=0.0, 0.2, 0.4, 0.6$, and 0.8) were synthesized by the sol–gel auto-combustion method[38], [39], [40], [41], [42]. Analytical-grade magnesium nitrate hexahydrate [$\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$], manganese nitrate

tetrahydrate $[\text{Mn}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}]$, cobalt nitrate hexahydrate $[\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}]$, iron nitrate nonahydrate $[\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}]$, and citric acid monohydrate $(\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O})$ were used as starting materials without further purification. Double-distilled water was employed as the solvent throughout the synthesis process. The required stoichiometric amounts of magnesium nitrate, manganese nitrate, cobalt nitrate, and iron nitrate corresponding to the composition $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ were separately dissolved in distilled water and subsequently mixed under continuous magnetic stirring to obtain a homogeneous solution. Citric acid was then added as a chelating agent. The solution was stirred continuously at 80–90 °C until complete dissolution of the reactants was achieved. The pH of the solution was adjusted to approximately 7–8 using aqueous ammonia solution. Continuous stirring and heating promoted complexation between the metal ions and citric acid, resulting in the formation of a transparent and homogeneous sol. Upon further evaporation of water, the sol gradually transformed into a highly viscous gel. The gel was heated at approximately 180–200 °C, leading to a self-sustained auto-combustion reaction. The combustion process produced a voluminous and fluffy precursor powder owing to the rapid release of gaseous products such as CO_2 , N_2 , and H_2O vapor. The as-burnt powder was lightly ground using an agate mortar and pestle to obtain a fine powder. the nanocrystalline powders were ground thoroughly to obtain homogeneous nanocrystalline $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrite samples suitable for structural, morphological, optical, electrical, and magnetic characterizations.



Figure:1. Schematic illustration of the sol–gel synthesis process for nanocrystalline A series of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ (where $x=0.0, 0.2, 0.4, 0.6$, and 0.8).

3. Results and Discussion

3.1. Morphological Study (FE-SEM and AFM)

The surface morphology and microstructural characteristics of the $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ (where $x=0.0, 0.2, 0.4, 0.6$, and 0.8) nanocrystalline ferrites were investigated using Field Emission Scanning Electron Microscopy (FESEM) and Atomic Force Microscopy (AFM). The FESEM micrographs reveal the formation of nearly spherical to irregularly shaped nanoparticles distributed throughout the surface. A noticeable degree of particle agglomeration is observed in all compositions, which is a common feature of ferrite nanoparticles synthesized by the sol–gel auto-combustion method. The agglomeration can be attributed to the high surface energy and magnetic dipole–dipole interactions among the nanoparticles[43], [44].

For the sample corresponding to figure 2 (a), the microstructure consists of relatively fine and uniformly distributed grains with moderate agglomeration. As the Mn concentration increases as shown in figure 2 (b and c), the particles tend to form larger agglomerated clusters with a denser packing arrangement. This behavior suggests enhanced grain growth and stronger interparticle interactions due to cation substitution. The sample represented by figure 2 (d) exhibits comparatively finer grains distributed over the surface, indicating that Mn substitution influences the nucleation and growth kinetics during ferrite formation. Overall, the FESEM observations confirm the nanocrystalline nature of the synthesized ferrites and indicate that Mn incorporation significantly affects grain morphology and particle aggregation.

The AFM images further support the FESEM observations by providing information about the surface topography and roughness of the samples. The AFM micrograph in figure 3 (a) shows a relatively smooth and homogeneous surface with low height variations of sample $x=0.2$. In contrast, figure 3 (b) exhibits pronounced ridge-like structures and increased surface irregularities, indicating a higher surface roughness. The figure 3 (c) displays a heterogeneous topography with interconnected surface features and distinct height fluctuations, suggesting non-uniform grain growth. The AFM image of figure 3 (d) reveals larger surface protrusions and clustered grains, which may result from particle coalescence during calcination.

The progressive changes in surface texture observed in the AFM images indicate that Mn^{2+} substitution modifies the growth behavior of crystallites and affects the surface morphology of the ferrite nanoparticles. The combined FESEM and AFM analyses demonstrate that all samples possess nanostructured morphology with varying degrees of agglomeration, grain connectivity, and surface roughness. These morphological variations are expected to influence the magnetic, optical, and electrical properties of the

$\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites by altering grain boundary density, surface defects, and interparticle interactions.

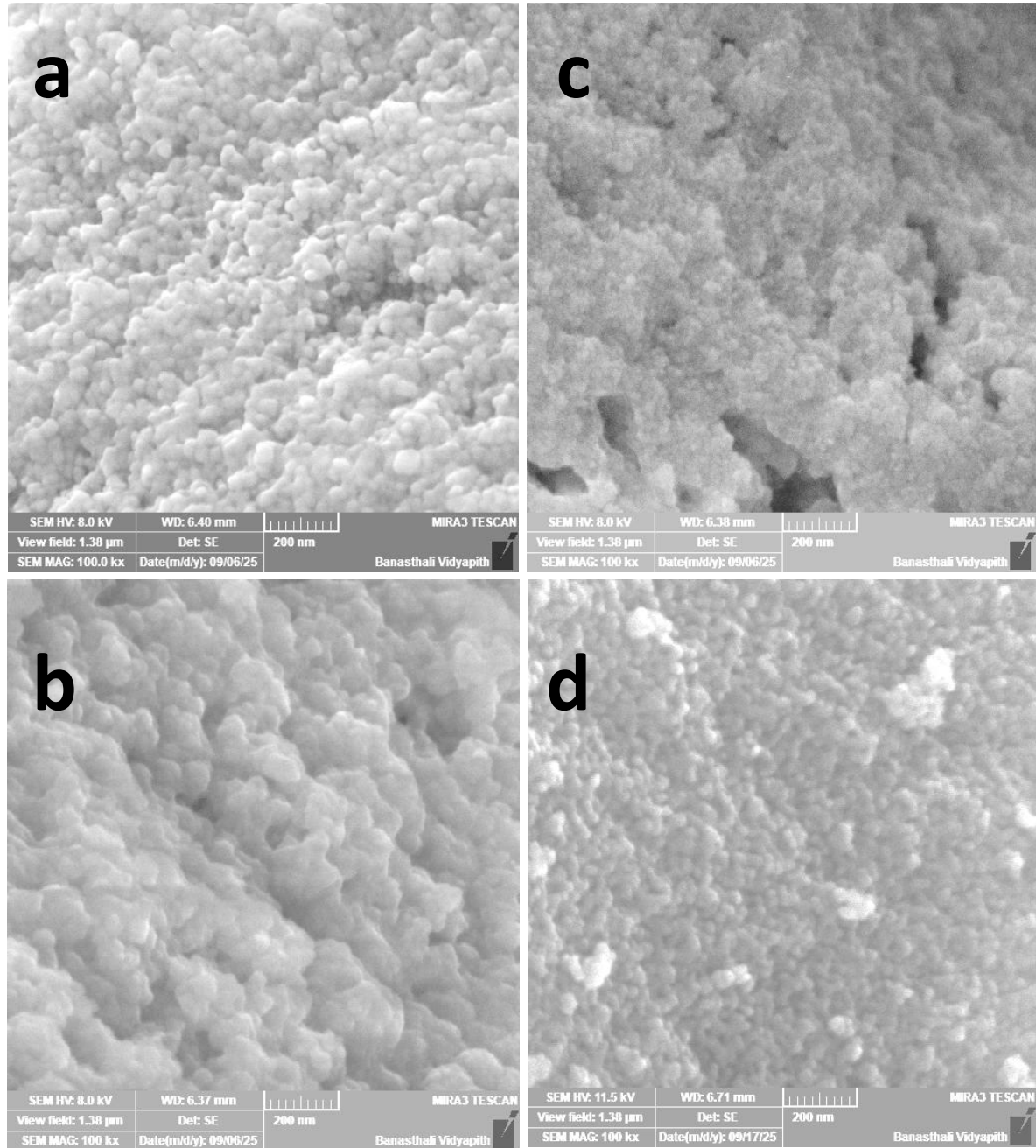


Figure:2. FESEM micrographs of nanocrystalline $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites with compositions (a) $x = 0.0$, (b) $x = 0.2$, (c) $x = 0.4$, (d) $x = 0.6$, and (e) $x = 0.8$.

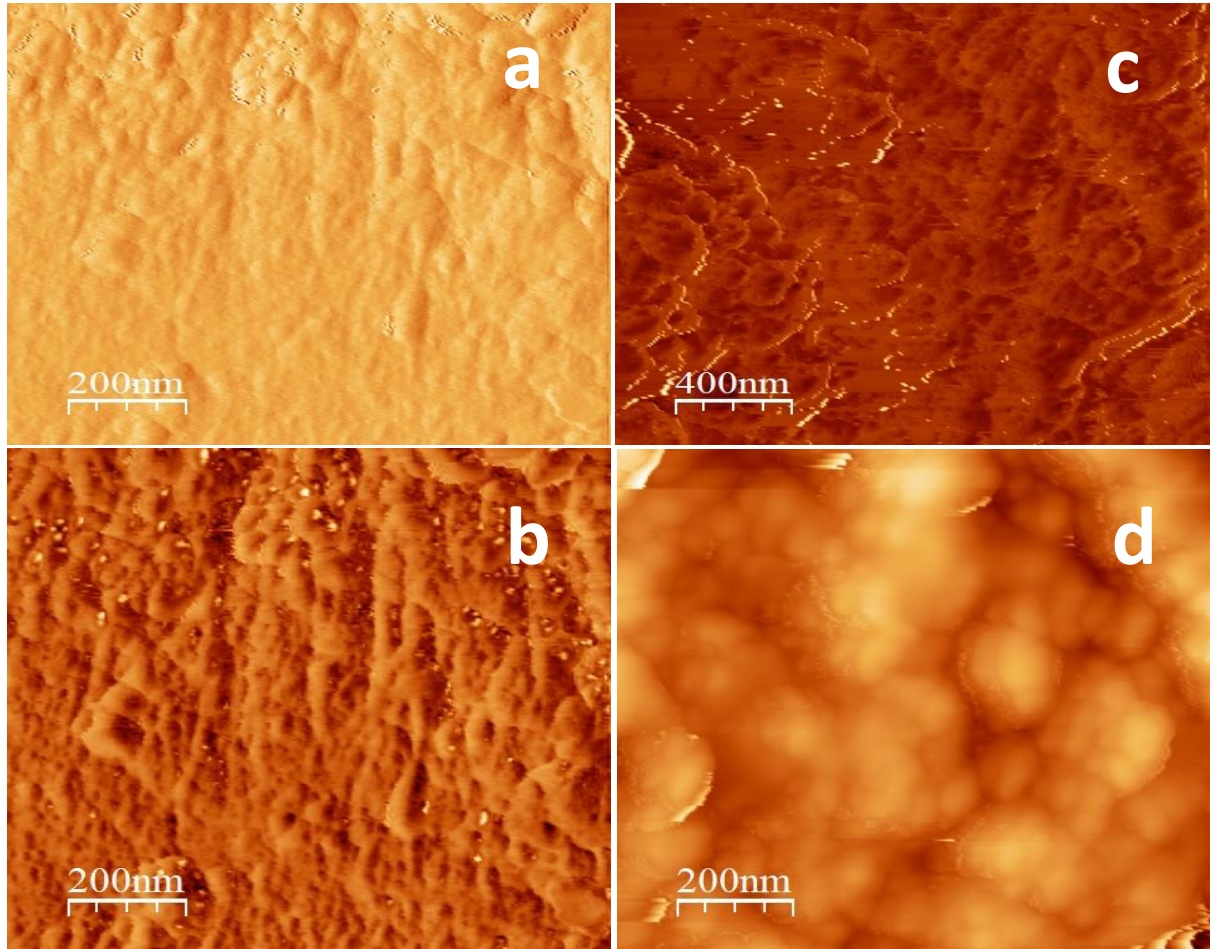


Figure:3. AFM images of nanocrystalline $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites corresponding to compositions (a) $x = 0.0$, (b) $x = 0.2$, (c) $x = 0.4$, (d) $x = 0.6$, and (e) $x = 0.8$.

3.2. X-ray diffraction analysis-

3.2.1. XRD analysis by Rietveld Refinement

All the synthesized samples of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ (where $x=0.0, 0.2, 0.4, 0.6$, and 0.8) were subjected to Rietveld refinement using the standard ICDD card No. 00-022-1086. The XRD analysis confirmed that the prepared samples exhibit a nanostructured nature and crystallize in the cubic spinel phase[45], [46], [47], [48], [49]. The best agreement between the experimental and calculated intensity profiles was obtained through least-squares fitting. The Rietveld refinement patterns along with the crystallographic structures of all samples are presented in **Figure 4**. During the refinement process, structural parameters such as atomic positions, site

occupancy, lattice constants, profile shape, asymmetry, preferred orientation, and thermal (isothermal) parameters were refined systematically.

The broadening of the diffraction peaks observed in the XRD patterns suggests the existence of microstructural strain along with reduced crystallite size[50], [51], [52]. Therefore, the (311) diffraction plane was selected for the estimation of crystallite size. The crystallite sizes of the prepared samples were calculated using the Debye–Scherrer equation.

$$D = 0.9\lambda/\beta\cos\theta \quad 1$$

Here, D represents the size of the crystallite, λ is represents the wavelength of X-ray, β is represents the FWHM, and θ is represents Bragg's angle. **Table 1** indicates that the crystallite size has been found between **20 and 50 nm**. The XRD data analysis showed that the lattice parameters reduced **from 8.43Å to 8.36Å** when the nickel concentration increases.

Table 1. Structural parameters of synthesized $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ (Where $x=0.0, 0.2, 0.4, 0.6$, and 0.8).

S. No.	Composition (x)	Peak position	Crystallite Size(D)nm	Lattice parameter [Å]	Volume [Å ³]	X-Ray density(g/cm ³)
1.	0.0	35.57	50	8.369	586.33	5.32
2.	0.2	35.60	36	8.362	584.88	5.30
3.	0.4	35.54	32	8.376	587.74	5.27
4.	0.6	35.46	23	8.394	591.58	5.22
5.	0.8	35.28	20	8.435	600.33	5.11

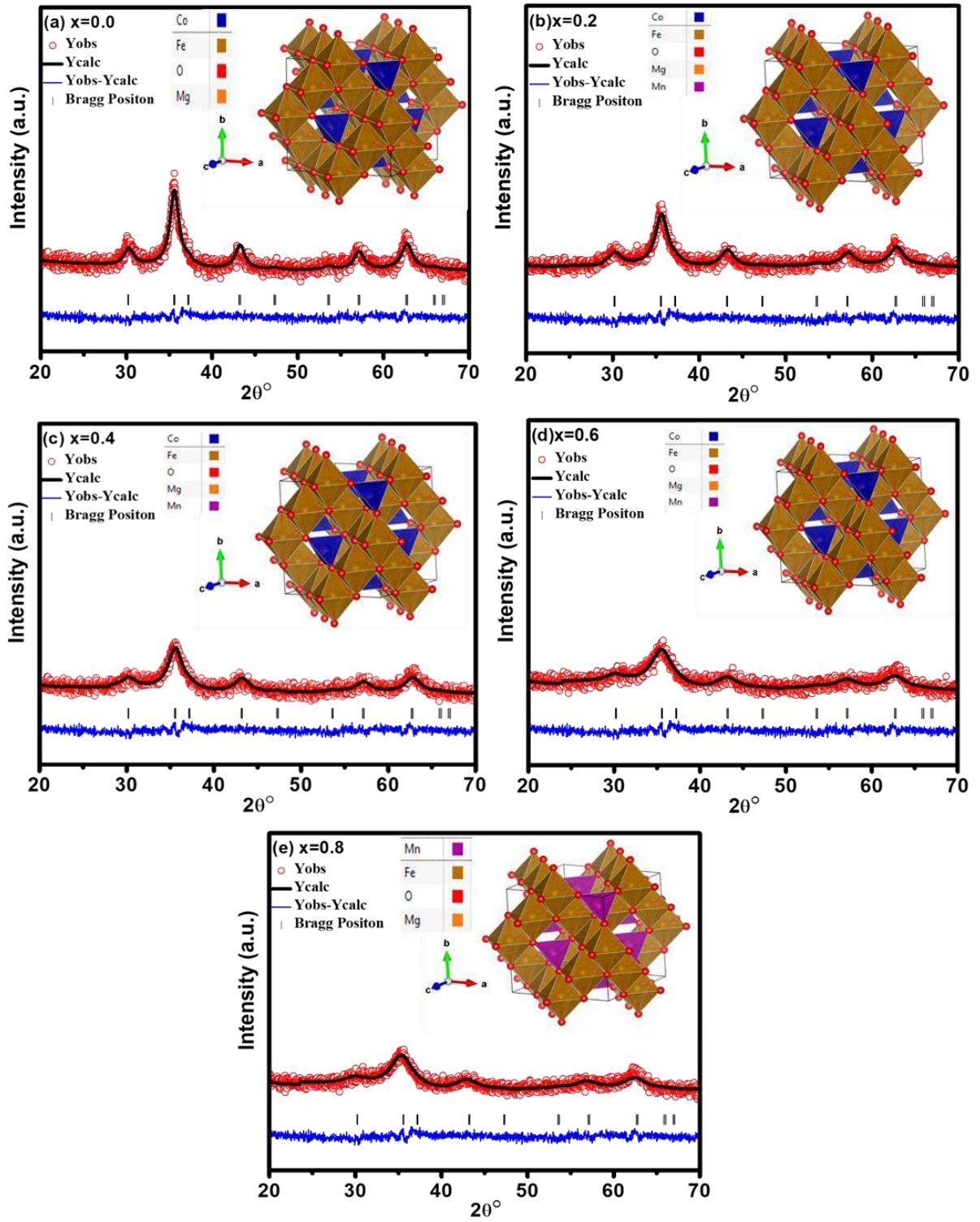


Figure:4. Rietveld-refined X-ray diffraction (XRD) patterns along with the corresponding crystal structures of nanocrystalline $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites for (a) $x = 0.0$, (b) $x = 0.2$, (c) $x = 0.4$, (d) $x = 0.6$, and (e) $x = 0.8$

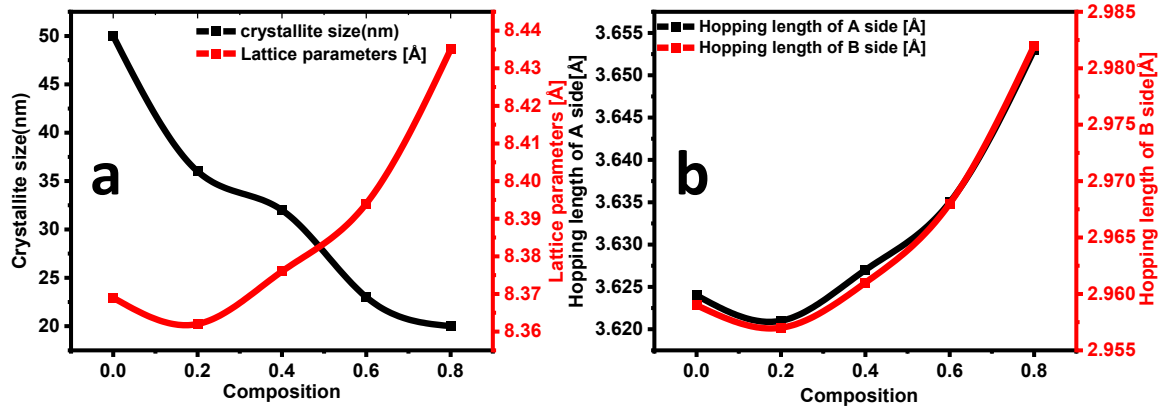


Figure:5. Variation of (a) crystallite size and lattice parameter, and (b) tetrahedral (L_a) and octahedral (L_β) hopping lengths as a function of Mn concentration in $Mg_{0.2}Mn_xCo_{0.8-x}Fe_2O_4$ nanocrystalline ferrites.

Figure 5 (a) illustrates the variation of crystallite size and lattice parameter with Mn concentration (x), while Figure (b) presents the corresponding changes in the tetrahedral (L_a) and octahedral (L_β) hopping lengths for $Mg_{0.2}Mn_xCo_{0.8-x}Fe_2O_4$ ferrites. The crystallite size (black line) decreases throughout the composition range. The absence of significant fluctuations suggests that all samples experienced similar nucleation and grain growth conditions during calcination. In contrast, the lattice parameter (red line) exhibits a slight decrease from $x = 0.0$ to $x = 0.2$, followed by a continuous increase with further Mn substitution, reaching its maximum value at $x = 0.8$. The initial decrease may arise from local lattice rearrangement and redistribution of cations between tetrahedral (A) and octahedral (B) sites. As the Mn content increases further, the lattice expands due to the gradual replacement of Co^{2+} ions by Mn^{2+} ions. Since the ionic radius of Mn^{2+} (0.80 Å) is larger than that of Co^{2+} (0.74 Å), incorporation of Mn^{2+} causes an increase in the interionic spacing and consequently enlarges the unit cell dimensions. This behavior follows Vegard's law and confirms the successful incorporation of Mn ions into the spinel lattice without disturbing the cubic crystal structure.

The hopping lengths at tetrahedral (L_a) and octahedral (L_β) sites are related to the lattice parameter through:

$$L_A = \frac{a\sqrt{3}}{4} \quad 5$$

$$L_B = \frac{a\sqrt{2}}{4} \quad 6$$

where a is the lattice parameter.

Since both hopping lengths are directly proportional to the lattice parameter, their variation closely follows the trend of lattice expansion. The values of both L_a and L_β show a slight decrease at $x = 0.2$ followed by a systematic increase with further Mn substitution. The overall increase in hopping lengths indicates an increase in the interionic distances between magnetic ions occupying A and B sites.

The increase in L_a and L_β suggests a larger separation between magnetic ions, which can influence the super-exchange interactions responsible for ferrimagnetic ordering. As the hopping lengths increase, the overlap between $\text{Fe}^{3+}\text{--O}^{2-}\text{--Fe}^{3+}$ and $\text{Fe}^{3+}\text{--O}^{2-}\text{--Mn}^{2+}$ orbitals may decrease slightly, modifying the A–B exchange interaction strength. Simultaneously, the introduction of Mn^{2+} ions, which possess a higher magnetic moment (5 μB) than Co^{2+} ions (3 μB), can compensate for the increased interionic distance and contribute to enhanced magnetic ordering.

The observed increase in lattice parameter with Mn substitution is attributed to the larger ionic radius of Mn^{2+} compared to Co^{2+} ions. Consequently, the hopping lengths at both tetrahedral and octahedral sites increase with increasing Mn content, reflecting the expansion of the spinel lattice. The initial decrease followed by an increase in crystallite size suggests a competition between strain-induced growth inhibition and thermally driven grain growth. The enlargement of hopping lengths modifies the cation–anion–cation distances and is expected to influence the super-exchange interactions, thereby affecting the magnetic and electrical

properties of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites. The systematic variation of these structural parameters confirms the successful incorporation of Mn ions into the spinel framework and highlights their significant role in tailoring the physical properties of the ferrite system

The structural parameters of the $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ nanoferrites were evaluated from the XRD data. The lattice parameter exhibited a slight decrease at lower Mn concentrations followed by a gradual increase with further Mn substitution, which can be attributed to the difference in ionic radii between Co^{2+} and Mn^{2+} ions and the consequent modification of the spinel lattice.

The X-ray density (ρ_x) of the synthesized ferrites was calculated using the following relation

$$X - ray\ density = \frac{ZM}{Na^3} \quad 2$$

where Z is the number of formula units per unit cell ($Z = 8$ for spinel ferrites), M is the molecular weight of the composition, N_a is Avogadro's number, and a is the lattice parameter. The variation in X-ray density with Mn concentration is governed by changes in both the molecular weight and the unit-cell volume of the ferrites. The dislocation density (σ), which provides information regarding the concentration of crystallographic defects within the material, was estimated using the relation

$$\sigma_x = \frac{1}{D^2} \quad 3$$

where D represents the average crystallite size obtained from the Debye–Scherrer equation. A lower dislocation density indicates improved crystallinity and reduced lattice imperfections in the synthesized ferrites.

Furthermore, the Williamson–Hall (W–H) method was employed to determine the lattice strain (ϵ) and to separate the contributions of crystallite size and microstrain to the XRD peak broadening. The Williamson–Hall equation is expressed as

$$\beta \cos \theta = 0.9 \lambda / D + 4 \varepsilon \sin \theta$$

4

where λ is the wavelength of the Cu-K α radiation, β is the full width at half maximum (FWHM) of the diffraction peak, D is the crystallite size, and ε represents the microstrain present in the crystal lattice. The W–H plots were constructed by plotting $\beta \cos \theta$ against $4 \sin \theta$, as shown in **Figure 6**. The slope of the linear fit yields the lattice strain, while the intercept provides the crystallite size contribution. In the present study, the positive slope of the Williamson–Hall plots indicates the presence of tensile strain within the crystal lattice. The calculated structural parameters, including crystallite size, lattice parameter, X-ray density, dislocation density, and lattice strain, are summarized in table 2.

Table 2. Structural parameters of synthesized $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ (Where $x=0.0, 0.2, 0.4, 0.6$, and 0.8).

S. No.	Composition (x)	Peak position	Dislocation density ($\delta \cdot 10^{14}$)	Specific surface area(m^2/g)	L_A [Å]	L_B [Å]
1.	0.0	35.57	4.53	24.0	3.624	2.959
2.	0.2	35.60	4.16	23.1	3.621	2.957
3.	0.4	35.54	4.53	24.2	3.627	2.961
4.	0.6	35.46	4.00	23.0	3.635	2.968
5.	0.8	35.28	3.70	22.6	3.653	2.982

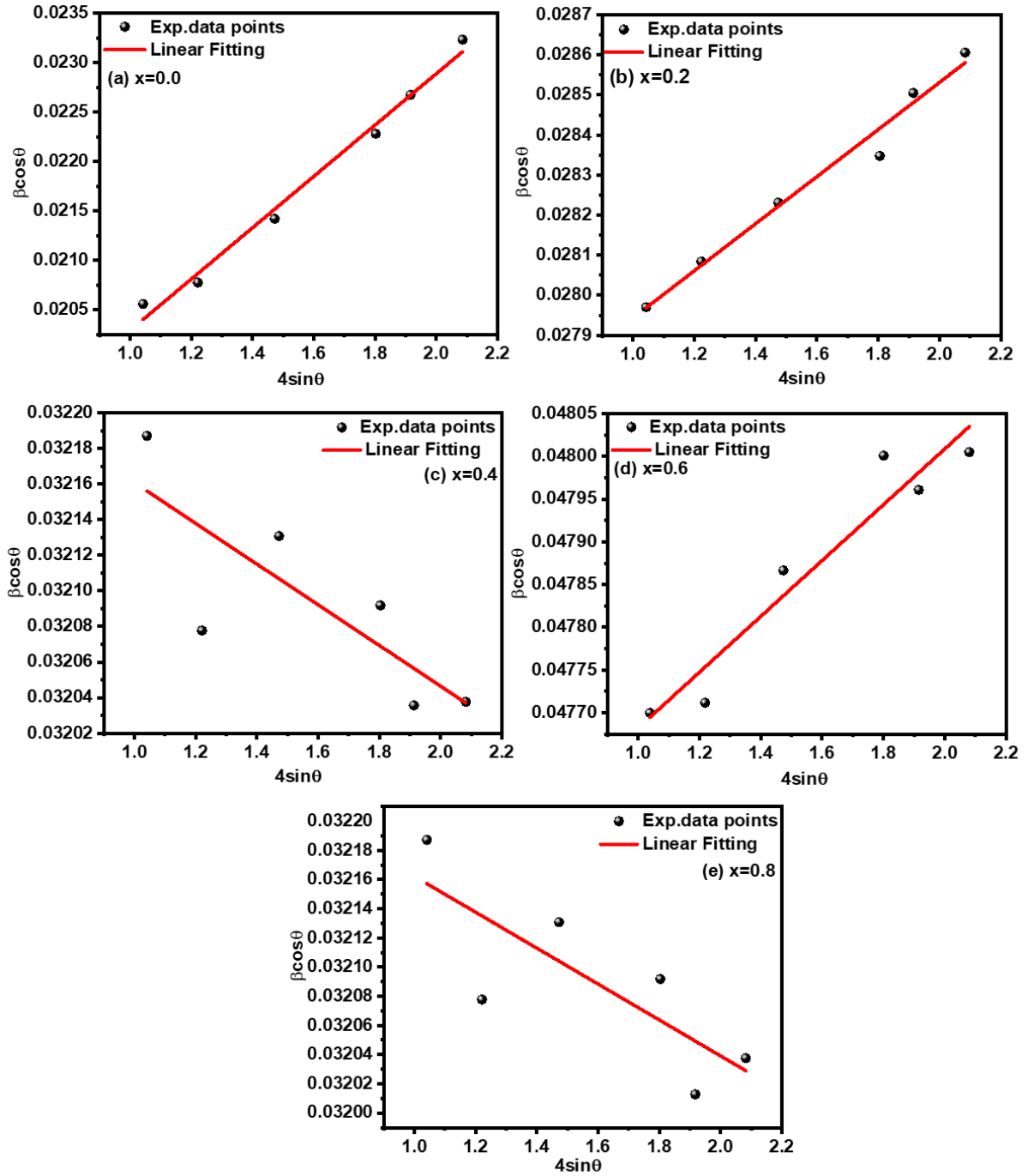


Figure:6. W-H plot of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ (where $x=0.0, 0.2, 0.4, 0.6$, and 0.8) of nanocrystalline ferrite samples.

3.3. Magnetic Studies

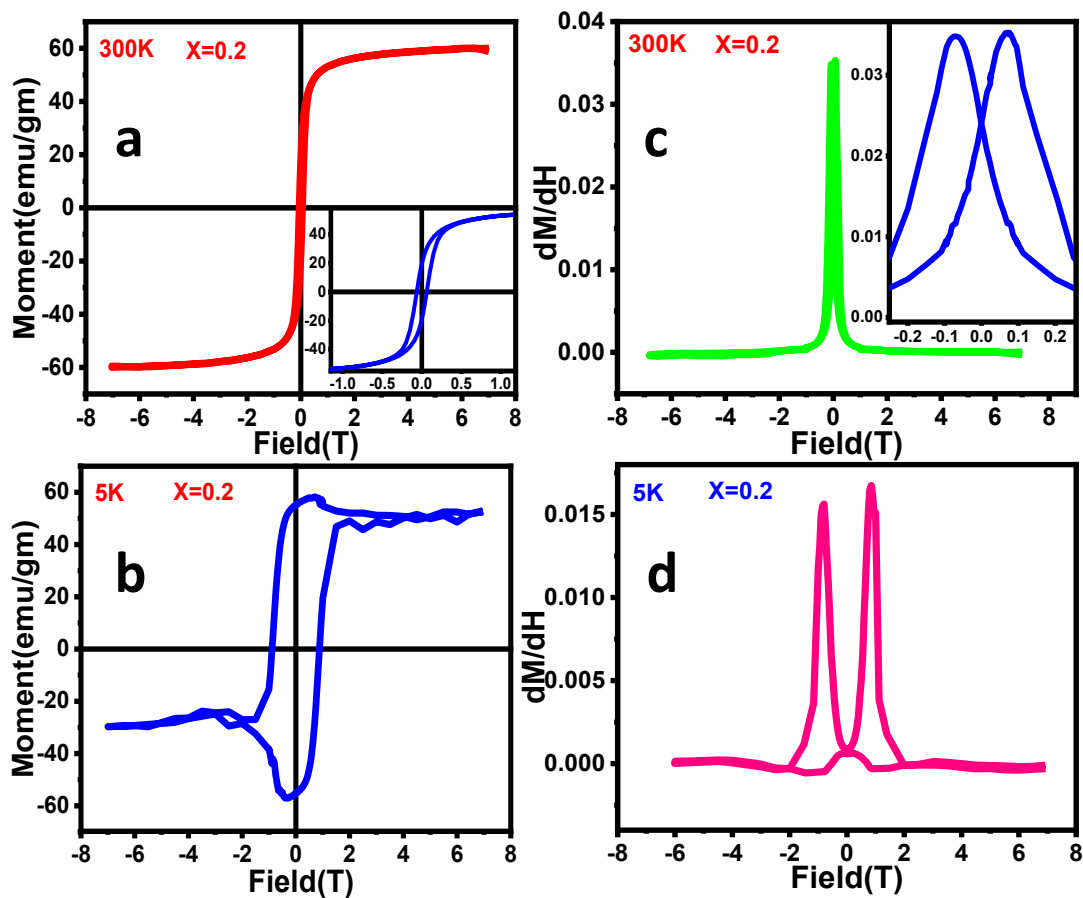


Figure:7. Magnetic hysteresis loops of $\text{Mg}_{0.2}\text{Mn}_{0.2}\text{Co}_{0.6}\text{Fe}_2\text{O}_4$ ferrite measured at (a) 300 K and (b) 5 K, alongwith the corresponding differential susceptibility curves (dM/dH) at (c) 300 K and (d) 5 K.

Figure 7 presents the magnetic hysteresis loops and their corresponding first-order derivatives (dM/dH) for the $\text{Mg}_{0.2}\text{Mn}_{0.2}\text{Co}_{0.6}\text{Fe}_2\text{O}_4$ ferrite measured at 300 K and 5 K. The hysteresis loop measured at 300 K (Figure 7 (a)) exhibits a narrow S-shaped profile with relatively low coercivity and remanent magnetization. The magnetization increases rapidly at low magnetic fields and gradually approaches saturation at higher fields. Such behavior is characteristic of a soft ferrimagnetic material in which domain wall motion and spin rotation occur with relatively low energy barriers. The narrow hysteresis width indicates that thermal energy at room temperature assists the magnetization reversal process, reducing magnetic anisotropy effects. The small coercive field suggests that magnetic domains can be reoriented

easily under an external magnetic field. The approach to saturation at moderate fields confirms the existence of long-range ferrimagnetic ordering arising from super-exchange interactions between magnetic ions occupying tetrahedral (A) and octahedral (B) sites.

At 5 K (Figure 7(b)), the hysteresis loop becomes significantly wider and more rectangular compared with the room-temperature loop. The coercivity and remanent magnetization increase substantially, indicating enhanced magnetic hardness at low temperature. The increase in coercivity can be attributed to the suppression of thermal fluctuations and the strengthening of magnetocrystalline anisotropy. At low temperatures, magnetic moments become more strongly pinned, making magnetization reversal more difficult. The larger loop area further indicates increased magnetic energy loss and stronger irreversible magnetization processes. The nearly saturated magnetization at high fields suggests that ferrimagnetic ordering remains dominant; however, the increased coercivity reflects a greater resistance to domain wall motion. Such behavior is commonly observed in cobalt-containing ferrites due to the strong anisotropic contribution of Co^{2+} ions.

The first derivative of the magnetization with respect to magnetic field (dM/dH) provides valuable information about magnetic reversal mechanisms, coercive field distribution, and magnetic phase homogeneity. Figure 7(c) shows the derivative curve measured at 300 K. A single sharp symmetric peak centered near zero field is observed. The narrow peak width indicates a relatively small distribution of switching fields and a homogeneous magnetic phase. The sharp maximum corresponds to the field region where magnetization changes most rapidly, reflecting efficient domain wall motion and low anisotropy barriers. The absence of multiple peaks suggests that the sample behaves predominantly as a single ferrimagnetic phase without significant magnetic phase separation. The narrow differential susceptibility peak is consistent with the low coercivity observed in the M–H loop and confirms soft magnetic behavior at room temperature.

The derivative curve obtained at 5 K (Figure 7(d)) exhibits two well-defined symmetric peaks located on either side of zero field. These peaks correspond closely to the positive and negative coercive fields of the hysteresis loop. The separation between the peaks increases significantly compared with the room-temperature case, directly reflecting the increase in coercivity at low temperature. The broader peak structure indicates a wider distribution of magnetic anisotropy energies and switching fields. The appearance of distinct double peaks suggests that magnetization reversal occurs through a combination of domain wall pinning and coherent spin rotation processes. At low temperatures, thermal activation is greatly reduced, causing magnetic moments to overcome larger energy barriers before reversal occurs.

The comparison between 300 K and 5 K measurements clearly demonstrates a strong temperature dependence of the magnetic properties of $\text{Mg}_{0.2}\text{Mn}_{0.2}\text{Co}_{0.6}\text{Fe}_2\text{O}_4$ ferrite: At **300 K**, the sample exhibits soft ferrimagnetic behavior characterized by low coercivity, narrow hysteresis, and a single sharp dM/dH peak. At **5 K**, thermal fluctuations are suppressed, leading to enhanced magnetic anisotropy, increased coercivity, larger remanence, and a broader double-peaked dM/dH profile. The derivative analysis confirms that the magnetic reversal mechanism evolves from relatively easy domain wall motion at room temperature to strongly pinned magnetic moments and enhanced anisotropy-controlled reversal at low temperature. The observed behavior highlights the important role of Co^{2+} -induced magnetocrystalline anisotropy and the temperature-dependent A–B super-exchange interactions in governing the magnetic response of $\text{Mg}_{0.2}\text{Mn}_{0.2}\text{Co}_{0.6}\text{Fe}_2\text{O}_4$ ferrites.

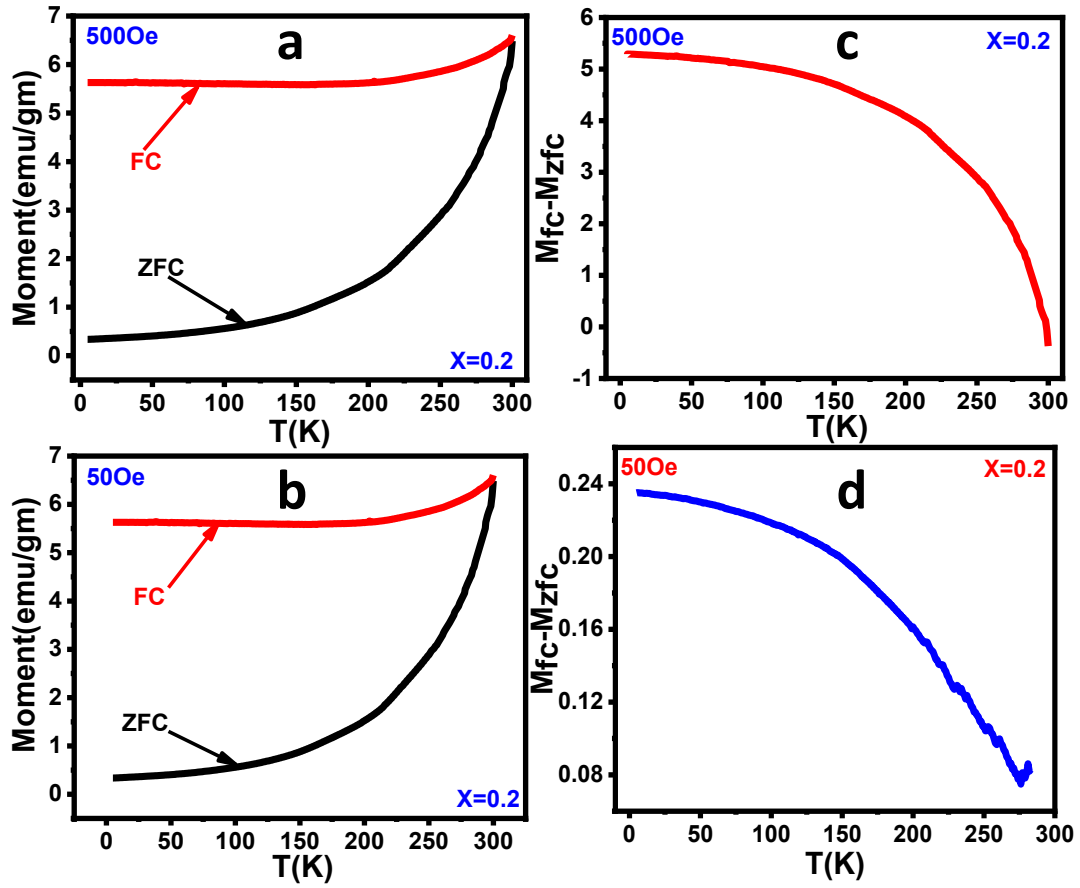


Figure:8. ZFC and FC magnetization curves of $\text{Mg}_{0.2}\text{Mn}_{0.2}\text{Co}_{0.6}\text{Fe}_2\text{O}_4$ ferrite measured under applied magnetic fields of (a) 500 Oe and (b) 50 Oe. The corresponding thermomagnetic irreversibility, expressed as $\Delta M = M_{\text{FC}} - M_{\text{ZFC}}$, is shown in (c) for 500 Oe and (d) for 50 Oe.

Figure 8 shows the temperature dependence of magnetization measured under zero-field-cooled (ZFC) and field-cooled (FC) conditions for the $\text{Mg}_{0.2}\text{Mn}_{0.2}\text{Co}_{0.6}\text{Fe}_2\text{O}_4$ ferrite at applied magnetic fields of 500 Oe and 50 Oe. The corresponding differences between FC and ZFC magnetization, $\Delta M = M_{\text{FC}} - M_{\text{ZFC}}$, are presented in Figures 8(c) and 8(d), respectively.

Figure 8(a) displays the ZFC and FC magnetization curves measured under an applied field of 500 Oe. The FC magnetization remains significantly higher than the ZFC magnetization throughout the entire temperature range, indicating a pronounced magnetic irreversibility. The ZFC curve increases gradually with temperature and then rises sharply at higher temperatures,

whereas the FC curve remains nearly constant before exhibiting a slight increase near the upper temperature limit. The absence of a distinct maximum in the ZFC curve suggests that the blocking temperature (T_B) is higher than the measured temperature range or that the system possesses a broad distribution of energy barriers associated with particle size and magnetic anisotropy. The large separation between the FC and ZFC branches indicates strong magnetocrystalline anisotropy and significant domain-wall pinning effects, which are commonly observed in cobalt-containing ferrites. Such behavior is characteristic of a ferrimagnetic system exhibiting thermomagnetic irreversibility rather than ideal superparamagnetic relaxation[53], [54], [55].

The FC and ZFC curves measured under 50 Oe exhibit similar qualitative behavior but with a larger separation between the two branches. At this lower magnetic field, the magnetic moments experience weaker alignment by the external field, allowing anisotropy effects and interparticle interactions to dominate the magnetic response. The enhanced bifurcation between FC and ZFC curves under 50 Oe suggests stronger magnetic frustration, spin freezing, or domain-wall pinning effects. The persistence of irreversibility throughout the measured temperature range indicates that the sample remains magnetically ordered and does not undergo a transition to a fully superparamagnetic state.

Figure 8(c) presents the temperature dependence of $\Delta M = M_{FC} - M_{ZFC}$ under 500 Oe. The difference remains positive over the entire temperature range, confirming that the FC magnetization is always greater than the ZFC magnetization. The nearly constant value of ΔM at low temperatures indicates strong magnetic freezing and stable ferrimagnetic ordering. As temperature increases, ΔM gradually decreases, suggesting that thermal energy assists the alignment of moments during the ZFC process and reduces magnetic irreversibility. The

reduction in ΔM reflects the progressive overcoming of anisotropy barriers and enhanced thermal activation of magnetic moments.

Figure 8(d) shows the corresponding $\Delta M = M_{FC} - M_{ZFC}$ variation measured under 50 Oe. The magnitude of ΔM is larger than that observed under 500 Oe, indicating stronger irreversibility at lower applied fields. The difference decreases continuously with increasing temperature, demonstrating that thermal fluctuations progressively reduce the separation between the FC and ZFC states. The stronger irreversibility under 50 Oe confirms that the external field is insufficient to overcome anisotropy energy barriers effectively. Consequently, the magnetic moments remain trapped in metastable states during the ZFC process, producing a larger FC–ZFC separation.

The absence of a clear blocking peak and the persistent FC–ZFC bifurcation indicate that $Mg_{0.2}Mn_{0.2}Co_{0.6}Fe_2O_4$ behaves predominantly as a ferrimagnetic nanoparticle system with strong anisotropy rather than a non-interacting superparamagnetic assembly.

The ZFC–FC measurements reveal pronounced thermomagnetic irreversibility in $Mg_{0.2}Mn_{0.2}Co_{0.6}Fe_2O_4$ ferrite under both 50 Oe and 500 Oe applied fields. The persistent bifurcation of the FC and ZFC curves over the entire temperature range confirms the existence of strong magnetic anisotropy, interparticle interactions, and ferrimagnetic ordering. The larger FC–ZFC separation observed at 50 Oe compared with 500 Oe indicates that lower magnetic fields are less effective in overcoming anisotropy barriers. The ΔM analysis further demonstrates that magnetic irreversibility decreases with increasing temperature due to thermally activated relaxation processes, while the absence of a distinct blocking temperature suggests a broad distribution of anisotropy energies and strongly interacting ferrimagnetic nanoparticles.

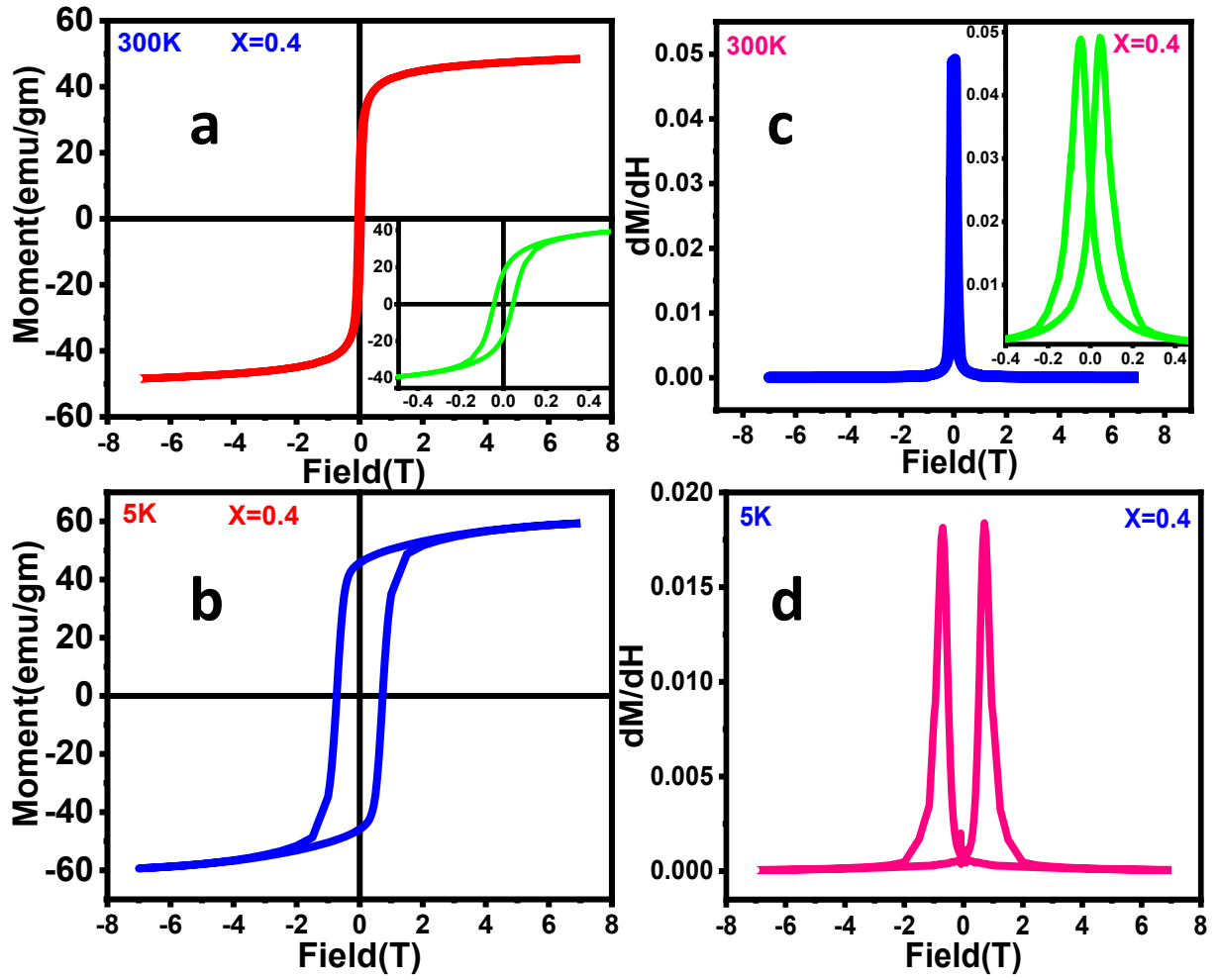


Figure:9. Magnetic hysteresis loops of $\text{Mg}_{0.2}\text{Mn}_{0.4}\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ferrite measured at (a) 300K and (b) 5K, together with the corresponding differential susceptibility curves (dM/dH) at (c) 300K and (d) 5K.

Figure 9 presents the magnetic hysteresis loops and corresponding differential susceptibility (dM/dH) curves of the $\text{Mg}_{0.2}\text{Mn}_{0.4}\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ferrite measured at 300K and 5K. The results provide insight into the temperature-dependent magnetic anisotropy, coercivity, and magnetization reversal mechanisms of the ferrite system.

Figure 9(a) shows the magnetic hysteresis loop measured at 300K. The loop exhibits a typical soft ferrimagnetic character with a narrow hysteresis width and rapid magnetization increase near the origin. The magnetization approaches saturation at relatively low magnetic fields, indicating efficient alignment of magnetic moments under the applied field. The small

coercive field and remanent magnetization suggest reduced magnetocrystalline anisotropy and easy domain-wall motion at room temperature. Thermal fluctuations facilitate spin rotation and domain reorientation, resulting in a soft magnetic response. The observed ferrimagnetic behavior originates from strong super-exchange interactions between magnetic ions occupying tetrahedral (A) and octahedral (B) sites of the spinel lattice. Compared with the $x = 0.2$ composition, the $x = 0.4$ sample exhibits a slightly steeper initial magnetization curve, indicating enhanced magnetic susceptibility and stronger exchange coupling due to increased Mn substitution[56].

The hysteresis loop recorded at 5 K (Figure 9(b)) is considerably broader than that measured at 300 K. A significant increase in coercivity, remanence, and hysteresis area is observed, demonstrating the strengthening of magnetic anisotropy at low temperature. At 5 K, thermal activation is greatly suppressed and magnetic moments become strongly pinned along preferred crystallographic directions. Consequently, higher magnetic fields are required to reverse the magnetization. The larger hysteresis area reflects increased magnetic energy loss associated with irreversible magnetization processes. The enhanced coercivity is characteristic of cobalt-containing ferrites, where Co^{2+} ions contribute significantly to magnetocrystalline anisotropy through spin–orbit coupling. The coexistence of Mn^{2+} and Co^{2+} ions modifies the A–B exchange interactions, leading to stronger ferrimagnetic ordering at low temperature[57], [58], [59], [60].

Figure 9(c) presents the first derivative of magnetization with respect to magnetic field measured at 300 K. The differential susceptibility curve exhibits a single sharp and symmetric peak centered around zero magnetic field. The narrow peak indicates a relatively small distribution of switching fields and confirms that the magnetization reversal process occurs primarily through domain-wall motion. The sharp maximum corresponds to the field region where magnetization changes most rapidly. The absence of additional peaks suggests magnetic homogeneity and the lack of secondary magnetic phases. The narrow peak width is consistent with the low coercivity observed in the hysteresis loop and confirms the soft magnetic nature of the sample at room temperature.

Figure 9(d) shows the dM/dH curve obtained at 5 K. In contrast to the room-temperature measurement, the derivative curve exhibits two distinct symmetric peaks positioned on either side of the zero-field axis. These peaks correspond to the positive and negative coercive fields and directly reflect the increased coercivity of the sample at low temperature. The larger

separation between the peaks indicates a significant enhancement in anisotropy energy barriers. The double-peak structure demonstrates that magnetization reversal occurs through a combination of domain-wall pinning and spin-rotation mechanisms. The sharpness of the peaks suggests relatively uniform switching fields, whereas the increased peak separation confirms stronger magnetic hardness at low temperature.

The magnetic behavior of $\text{Mg}_{0.2}\text{Mn}_{0.4}\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ is governed by the competition between thermal activation, magnetocrystalline anisotropy, and A–B super-exchange interactions. Increasing Mn substitution modifies the cation distribution within the spinel lattice and influences the exchange interactions between magnetic ions. At room temperature, these interactions produce a soft ferrimagnetic response, whereas at 5 K the anisotropy contribution becomes dominant, resulting in enhanced coercivity and magnetic hardness.

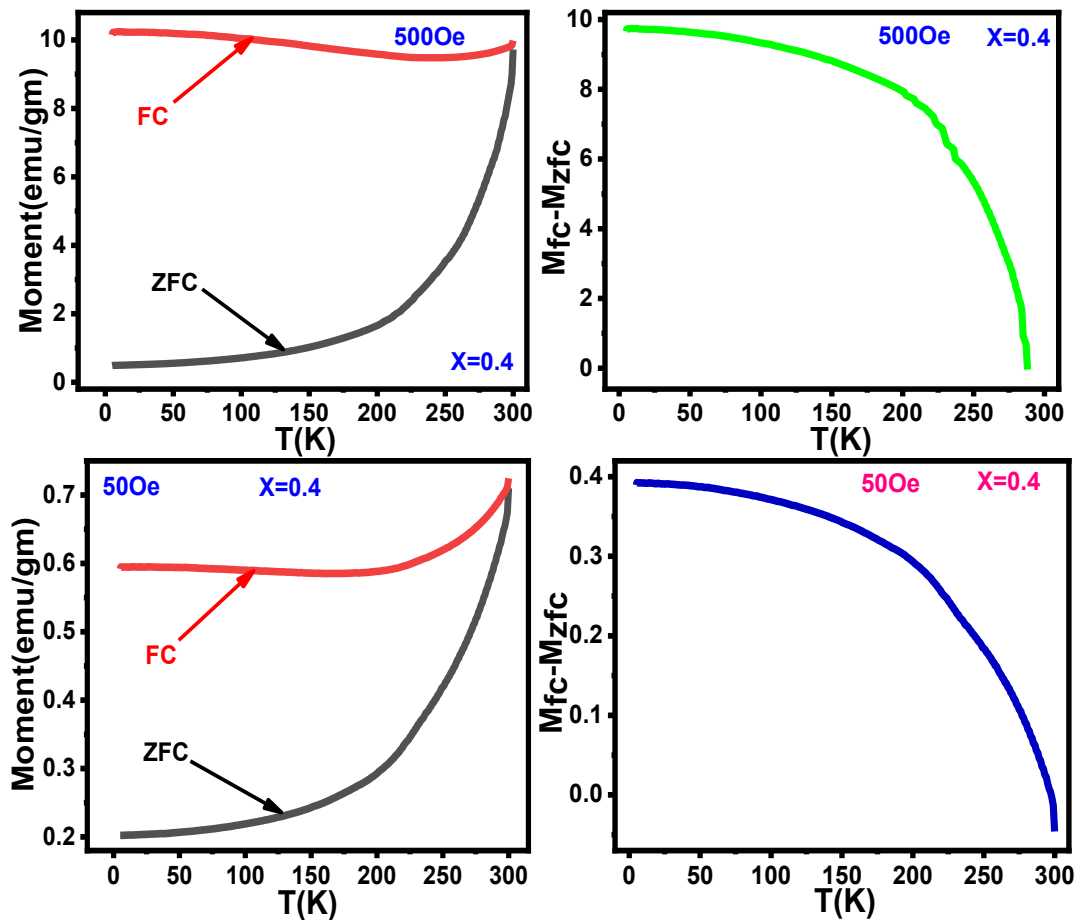


Figure:10. ZFC and FC magnetization curves of $\text{Mg}_{0.2}\text{Mn}_{0.4}\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ferrite measured under applied magnetic fields of (a) 500 Oe and (b) 50 Oe. The corresponding thermomagnetic irreversibility, expressed as $\Delta M = M_{FC} - M_{ZFC}$, is shown in (c) for 500 Oe and (d) for 50 Oe.

Figure 10(a) shows the ZFC and FC magnetization curves measured under an applied field of 500 Oe. The FC magnetization remains significantly higher than the ZFC magnetization throughout the entire temperature range, indicating pronounced magnetic irreversibility. The ZFC magnetization gradually increases with increasing temperature and rises sharply above approximately 200 K, while the FC curve remains nearly constant over a broad temperature range before exhibiting a slight increase near room temperature. The absence of a distinct maximum in the ZFC branch suggests that the blocking temperature (TB) lies above the measured temperature range or that the sample possesses a broad distribution of anisotropy energy barriers. The large separation between the FC and ZFC curves indicates strong magnetocrystalline anisotropy and substantial domain-wall pinning. Such behavior is typical of Co-containing ferrites, where the spin–orbit coupling of Co^{2+} ions contributes significantly to the anisotropy energy.

The ZFC and FC curves measured under 50 Oe (Figure 10(b)) exhibit a similar trend; however, the bifurcation between the two branches is more pronounced than that observed under 500 Oe. At lower magnetic fields, the external field is less effective in overcoming anisotropy barriers and aligning magnetic moments. Consequently, the magnetic moments remain trapped in metastable states during the ZFC process, resulting in lower ZFC magnetization and enhanced irreversibility. The persistent separation of FC and ZFC branches over the entire temperature range indicates that the ferrite remains magnetically ordered and does not exhibit complete superparamagnetic relaxation within the investigated temperature window.

Figure 10(c) shows the temperature dependence of ΔM measured under 500 Oe. The difference remains positive throughout the investigated temperature range, confirming that the FC magnetization is consistently larger than the ZFC magnetization. At low temperatures, ΔM attains its maximum value, indicating strong magnetic freezing and highly irreversible magnetization behavior. As temperature increases, ΔM gradually decreases, reflecting thermally activated relaxation of magnetic moments and enhanced domain-wall mobility. The smooth reduction of ΔM suggests a progressive weakening of magnetic anisotropy barriers with increasing temperature rather than an abrupt magnetic transition.

Figure 10(d) presents the ΔM curve obtained under 50 Oe. Similar to the 500 Oe measurement, ΔM decreases continuously with increasing temperature. However, the magnitude of ΔM remains comparatively larger over most of the temperature range, indicating stronger thermomagnetic irreversibility at lower applied fields. The larger FC–ZFC difference at 50 Oe confirms that the anisotropy energy barriers dominate the magnetic response when the external field is weak. The gradual decline of ΔM with increasing temperature demonstrates that thermal activation progressively assists the alignment of magnetic moments during the ZFC process. Near room temperature, ΔM approaches a minimum value, suggesting a significant reduction in magnetic irreversibility due to enhanced thermal fluctuations.

Compared with the $x = 0.2$ composition, the $x = 0.4$ sample exhibits a similar irreversible magnetic behavior, indicating that ferrimagnetic ordering remains dominant despite the increased Mn substitution. The coexistence of Mn^{2+} and Co^{2+} ions modifies the A–B superexchange interactions and contributes to the observed temperature-dependent magnetic response.

The pronounced bifurcation between the FC and ZFC magnetization curves and the non-zero ΔM values across the entire temperature range demonstrate that $Mg_{0.2}Mn_{0.4}Co_{0.4}Fe_2O_4$ possesses strong magnetic anisotropy and irreversible magnetization behavior. The larger FC–ZFC separation observed at 50 Oe compared with 500 Oe confirms that low applied fields are insufficient to overcome anisotropy energy barriers effectively. The continuous decrease of ΔM with increasing temperature reflects thermally assisted relaxation processes and enhanced domain-wall mobility. The absence of a well-defined blocking peak suggests a broad distribution of anisotropy energies and strong interparticle interactions, characteristic of ferrimagnetic nanocrystalline spinel ferrites.

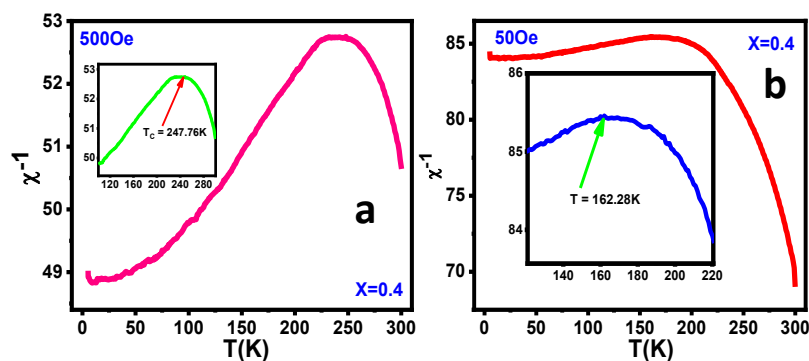


Figure:11. Temperature dependence of the inverse magnetic susceptibility (χ^{-1}) of $Mg_{0.2}Mn_{0.4}Co_{0.4}Fe_2O_4$ ferrite measured under applied magnetic fields of (a) 500 Oe and (b) 50 Oe

Figure 11(a) presents the χ^{-1} versus temperature curve measured under an applied field of 500 Oe. The inverse susceptibility exhibits a non-linear dependence on temperature throughout the investigated range, indicating a deviation from ideal Curie–Weiss paramagnetic behavior. At low temperatures, χ^{-1} remains relatively small due to the strong ferrimagnetic ordering and high magnetic susceptibility of the sample. With increasing temperature, χ^{-1} gradually increases, reflecting the progressive weakening of magnetic exchange interactions by thermal fluctuations. Near the higher temperature region, the χ^{-1} curve rises sharply, suggesting the approach toward a magnetic disordering regime. The inset χ –T curve exhibits a broad maximum around the intermediate temperature region. Such a broad peak is characteristic of ferrimagnetic nanoparticles possessing a distribution of anisotropy energies and interparticle interactions rather than a sharp magnetic phase transition. The broad nature of the peak indicates gradual thermal demagnetization associated with nanocrystalline ferrites. The deviation from linear Curie–Weiss behavior suggests the presence of short-range magnetic correlations above the magnetic ordering region, which are commonly observed in spinel ferrites due to strong A–B super-exchange interactions.

Figure 11(b) shows the inverse susceptibility measured under a lower magnetic field of 50 Oe. The overall behavior is similar to that observed at 500 Oe; however, the magnitude of χ^{-1} is reduced due to the enhanced susceptibility at lower applied fields. The χ^{-1} curve remains non-linear over the entire temperature range, confirming that magnetic interactions persist even at elevated temperatures. The lower applied field allows intrinsic magnetic interactions and anisotropy effects to dominate the magnetic response, leading to a stronger departure from ideal paramagnetic behavior. The inset χ –T curve exhibits a more pronounced maximum compared with the 500 Oe measurement. This peak corresponds to the temperature region where the competition between thermal agitation and magnetic ordering becomes significant. The

broad peak width further indicates a distribution of magnetic relaxation processes and anisotropy barriers.

The non-linear χ^{-1} -T behavior observed under both 500 Oe and 50 Oe confirms that $\text{Mg}_{0.2}\text{Mn}_{0.4}\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ does not behave as a simple paramagnet within the investigated temperature range.

The broad maxima observed in the χ -T curves suggest the presence of thermally activated magnetic relaxation and possible blocking phenomena associated with nanoparticle assemblies. The absence of a sharp divergence in χ^{-1} indicates that the magnetic transition is distributed over a wide temperature range, which is typical for nanostructured ferrites possessing particle-size distributions and surface-spin effects.

The temperature dependence of inverse magnetic susceptibility for $\text{Mg}_{0.2}\text{Mn}_{0.4}\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ reveals significant deviations from Curie-Weiss behavior under both 500 Oe and 50 Oe applied fields. The observed non-linearity of the χ^{-1} -T curves indicates the persistence of ferrimagnetic correlations and strong A-B super-exchange interactions over the entire temperature range. The broad maxima observed in the susceptibility curves reflect thermally activated magnetic relaxation processes and the influence of magnetic anisotropy, interparticle interactions, and surface-spin disorder. The more pronounced susceptibility peak under 50 Oe suggests that lower magnetic fields allow the intrinsic magnetic interactions of the ferrite nanoparticles to be observed more clearly. Overall, the χ^{-1} -T results confirm the presence of a strongly interacting ferrimagnetic nanoparticle system with distributed magnetic energy barriers and complex magnetic ordering behavior.

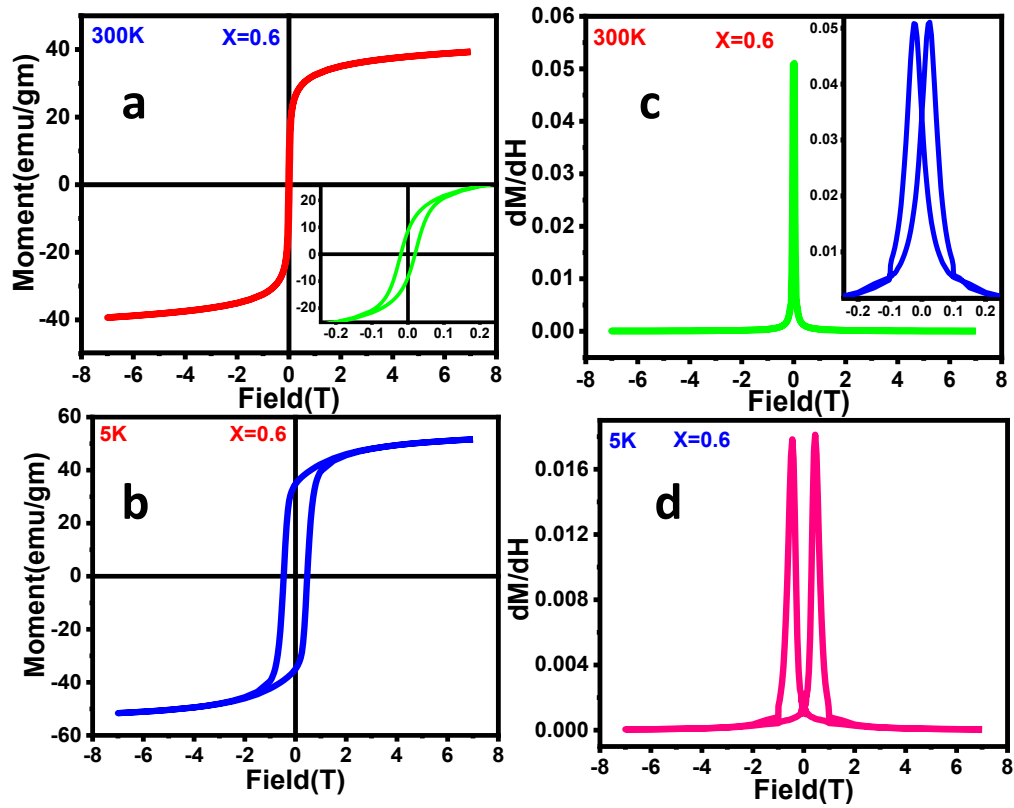


Figure: 12. Magnetic hysteresis loops of $\text{Mg}_{0.2}\text{Mn}_{0.6}\text{Co}_{0.2}\text{Fe}_2\text{O}_4$ ferrite measured at (a) 300 K and (b) 5 K, together with the corresponding differential susceptibility curves (dM/dH) at (c) 300 K and (d) 5 K.

Figure 12(a) displays the hysteresis loop measured at 300 K. The magnetization increases rapidly at low magnetic fields and gradually approaches saturation at higher fields, indicating a ferrimagnetic nature of the sample. The loop is relatively narrow with low coercivity and remanence, characteristic of soft magnetic behavior. The rapid magnetization rise near the origin indicates efficient domain-wall motion and easy spin alignment under the applied magnetic field. The reduced coercive field suggests that thermal energy at room temperature assists the reversal of magnetic domains by overcoming anisotropy barriers. The observed magnetic response confirms that ferrimagnetic interactions between ions occupying the tetrahedral (A) and octahedral (B) sites remain dominant at room temperature. Compared with the lower Mn compositions ($x = 0.2$ and $x = 0.4$), the $x = 0.6$ sample exhibits a steeper initial

magnetization and improved saturation tendency, indicating stronger A–B super-exchange interactions due to the increased Mn substitution.

The hysteresis loop recorded at 5 K [Figure 12(b)] becomes substantially broader than that observed at 300 K. A marked increase in coercivity, remanent magnetization, and hysteresis area is evident. At low temperature, thermal fluctuations are significantly suppressed and magnetic moments become strongly pinned along energetically favorable directions. Consequently, larger magnetic fields are required to reverse the magnetization. The enhanced coercivity indicates stronger magnetocrystalline anisotropy and increased resistance to domain-wall movement. The nearly rectangular shape of the hysteresis loop suggests stronger ferrimagnetic ordering and improved magnetic moment alignment. The larger loop area also reflects increased magnetic energy loss associated with irreversible magnetization processes.

Figure 12(c) shows the first derivative of magnetization with respect to magnetic field measured at 300 K. The differential susceptibility curve exhibits a sharp and symmetric peak centered near zero magnetic field. The narrow peak width indicates a small distribution of switching fields and relatively homogeneous magnetic behavior throughout the sample. The maximum of the dM/dH curve corresponds to the field region where magnetization changes most rapidly, which is associated primarily with domain-wall displacement. The absence of multiple peaks confirms that the sample behaves as a single ferrimagnetic phase without evidence of magnetic phase separation or secondary magnetic contributions.

Figure 12(d) presents the differential susceptibility measured at 5 K. In contrast to the room-temperature measurement, two well-defined peaks appear symmetrically on both sides of the zero-field axis. These peaks correspond to the positive and negative coercive fields and directly reflect the increased coercivity observed in the hysteresis loop. The larger peak separation indicates stronger anisotropy energy barriers and greater resistance to magnetization

reversal. The double-peaked structure demonstrates that the magnetization reversal process at 5 K is governed by domain-wall pinning and spin-rotation mechanisms. The sharpness of the peaks suggests relatively uniform switching fields, whereas their increased separation confirms enhanced magnetic hardness at low temperatures.

Comparison of the magnetic behavior at 300 K and 5 K reveals significant temperature-dependent changes: At **300 K**, thermal activation facilitates domain-wall motion and spin rotation, resulting in soft ferrimagnetic behavior with low coercivity. At **5 K**, thermal energy is insufficient to overcome anisotropy barriers, producing stronger magnetic irreversibility and larger coercivity. The transformation from a single central dM/dH peak at 300 K to a symmetric double-peak structure at 5 K confirms the increasing role of magnetocrystalline anisotropy in controlling magnetization reversal. The larger peak separation in the low-temperature derivative curve is a direct manifestation of enhanced coercive fields.

The magnetic behavior of $Mg_{0.2}Mn_{0.6}Co_{0.2}Fe_2O_4$ is governed by the interplay between ferrimagnetic A–B exchange interactions and temperature-dependent magnetic anisotropy. The increased Mn concentration modifies the cation distribution within the spinel lattice and strengthens the magnetic exchange interactions, while the remaining Co^{2+} ions continue to contribute significantly to anisotropy through spin–orbit coupling. At room temperature, thermal activation dominates the magnetization process, leading to soft magnetic characteristics. At 5 K, anisotropy effects become predominant, resulting in enhanced coercivity, increased remanence, stronger domain-wall pinning, and a more pronounced ferrimagnetic response.

The magnetic hysteresis measurements demonstrate that $Mg_{0.2}Mn_{0.6}Co_{0.2}Fe_2O_4$ exhibits ferrimagnetic behavior over the entire investigated temperature range. The room-temperature hysteresis loop is characterized by low coercivity and soft magnetic behavior, whereas the 5 K

loop displays enhanced coercivity and magnetic hardness due to the suppression of thermal fluctuations and strengthening of anisotropy effects. The differential susceptibility analysis corroborates these observations, revealing a transition from a single narrow peak at 300 K to a double-peaked structure at 5 K. These results indicate that magnetization reversal is dominated by domain-wall motion at room temperature and by anisotropy-controlled pinning mechanisms at low temperature, confirming the strong influence of temperature on the magnetic dynamics of $\text{Mg}_{0.2}\text{Mn}_{0.6}\text{Co}_{0.2}\text{Fe}_2\text{O}_4$ ferrite nanoparticles.

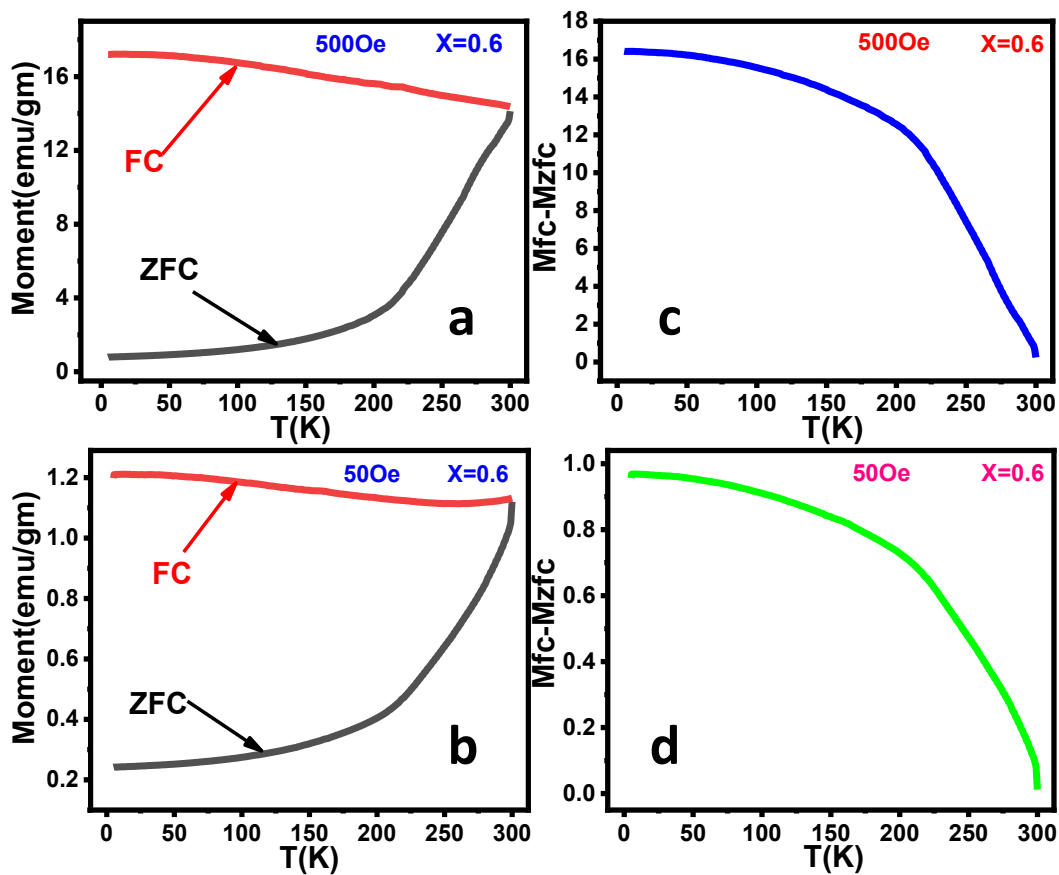


Figure:13. ZFC and FC magnetization curves of $\text{Mg}_{0.2}\text{Mn}_{0.6}\text{Co}_{0.2}\text{Fe}_2\text{O}_4$ ferrite measured under applied magnetic fields of (a) 500 Oe and (b) 50 Oe. The corresponding thermomagnetic irreversibility, expressed as $\Delta M = M_{FC} - M_{ZFC}$, is shown in (c) for 500 Oe and (d) for 50 Oe.

Figure 13(a) displays the ZFC and FC magnetization curves measured under an applied field of 500 Oe. The FC magnetization remains substantially higher than the ZFC magnetization throughout the investigated temperature range, indicating strong thermomagnetic irreversibility.

The ZFC curve exhibits a gradual increase with temperature and rises rapidly above approximately 200 K, while the FC curve shows a weak temperature dependence and decreases slightly with increasing temperature. The absence of a well-defined maximum in the ZFC branch suggests that the blocking temperature (TB) is higher than the maximum measurement temperature or that the sample contains a broad distribution of anisotropy energy barriers.

The persistent bifurcation between the FC and ZFC curves is characteristic of ferrimagnetic nanoparticles possessing strong magnetic anisotropy and interparticle interactions. Such behavior indicates that the magnetic moments remain partially frozen during the ZFC process and are unable to attain complete thermal equilibrium.

The ZFC and FC curves measured under 50 Oe [Figure 13(b)] exhibit similar behavior but with a larger separation between the FC and ZFC branches. Under the lower magnetic field, the external field is less effective in overcoming anisotropy barriers and aligning magnetic moments. Consequently, the ZFC magnetization remains significantly lower than the FC magnetization over the entire temperature range. The enhanced bifurcation confirms the existence of strong magnetic anisotropy and irreversible magnetization processes. The absence of a blocking peak further suggests that ferrimagnetic ordering dominates the magnetic response rather than superparamagnetic relaxation. The larger FC–ZFC separation at 50 Oe compared with 500 Oe demonstrates that the intrinsic magnetic interactions become more prominent when the applied field is reduced.

Figure 13(c) shows the temperature dependence of ΔM measured under 500 Oe. The difference remains positive throughout the temperature range, confirming that the FC state possesses a higher magnetization than the ZFC state. At low temperatures, ΔM reaches its maximum value due to strong spin freezing and magnetic anisotropy. As temperature increases, ΔM gradually decreases, indicating that thermal activation assists the alignment of magnetic moments during the ZFC process and reduces magnetic irreversibility. The smooth decrease in ΔM suggests a continuous thermal relaxation process rather than a sharp magnetic transition. This behavior is typical of nanocrystalline ferrites with distributed anisotropy energies and interparticle interactions.

Figure 13(d) presents the variation of ΔM under 50 Oe. Similar to the 500 Oe measurement, ΔM decreases continuously with increasing temperature; however, the magnitude of ΔM remains larger over most of the temperature range. The enhanced ΔM confirms stronger thermomagnetic irreversibility at lower applied fields. Since the external magnetic field is weaker, a larger fraction of magnetic moments remains trapped in metastable states during the ZFC process, resulting in greater separation between the FC and ZFC magnetizations. Near room temperature, ΔM decreases significantly due to thermally activated moment relaxation and improved magnetic moment alignment. Compared with the $x = 0.4$ composition, the $x = 0.6$ sample exhibits a similar FC–ZFC bifurcation behavior, indicating that ferrimagnetic ordering remains dominant despite the increased Mn substitution. The gradual reduction of ΔM with temperature reflects the progressive influence of thermal activation on magnetic moment relaxation.

The ZFC–FC magnetization measurements of $\text{Mg}_{0.2}\text{Mn}_{0.6}\text{Co}_{0.2}\text{Fe}_2\text{O}_4$ reveal pronounced thermomagnetic irreversibility under both 500 Oe and 50 Oe applied fields. The persistent separation between FC and ZFC curves over the entire temperature range indicates strong

magnetic anisotropy, interparticle interactions, and ferrimagnetic ordering. The larger FC–ZFC bifurcation observed at 50 Oe demonstrates that lower magnetic fields are less effective in overcoming anisotropy barriers, resulting in enhanced magnetic irreversibility. The ΔM analysis further confirms that thermal activation progressively reduces the difference between FC and ZFC states with increasing temperature. The absence of a distinct blocking temperature suggests a broad distribution of anisotropy energies and strong collective magnetic interactions, characteristic of nanocrystalline spinel ferrites.

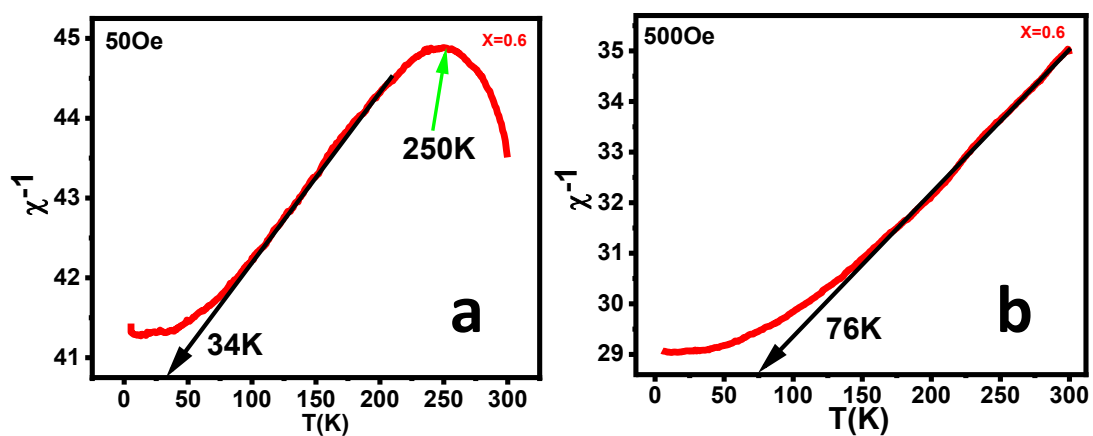


Figure:14. Temperature dependence of the inverse magnetic susceptibility (χ^{-1}) of $\text{Mg}_{0.2}\text{Mn}_{0.6}\text{Co}_{0.2}\text{Fe}_2\text{O}_4$ ferrite measured under applied magnetic fields of (a) 500 Oe and (b) 50 Oe.

Figure 14 presents the variation of inverse magnetic susceptibility (χ^{-1}) with temperature for the $x = 0.6$ composition measured under applied magnetic fields of 500 Oe and 50 Oe. The χ^{-1} – T curves provide important information regarding the magnetic interactions, magnetic ordering temperature, and deviation from ideal Curie–Weiss behavior.

The inverse susceptibility initially increases gradually with increasing temperature and follows an approximately linear trend over a broad temperature range. The linear region indicates that the sample obeys the Curie–Weiss law in the paramagnetic regime. A noticeable deviation from linearity appears at lower temperatures, indicating the onset of short-range magnetic correlations and ferrimagnetic ordering. The positive extrapolated Curie–Weiss

temperature suggests dominant ferromagnetic/ferrimagnetic exchange interactions between magnetic ions occupying the tetrahedral (A) and octahedral (B) sites.

The broad maximum observed near the high-temperature region indicates the magnetic transition from an ordered ferrimagnetic state to a paramagnetic state. This temperature can be associated with the Curie temperature (TC). The broad nature of the peak suggests a distribution of magnetic exchange interactions, which is commonly observed in nanocrystalline ferrites due to finite-size effects, cation disorder, and surface spin disorder.

The χ^{-1} -T curve measured under 50 Oe exhibits a similar overall trend but shows stronger curvature and greater deviation from ideal Curie-Weiss behavior. At low magnetic fields, the susceptibility becomes more sensitive to magnetic anisotropy, domain-wall pinning, and spin-cluster effects. The pronounced nonlinearity at lower temperatures indicates the presence of strong spin correlations and magnetic inhomogeneities. Compared with the 500 Oe measurement, the low-field data reveal magnetic fluctuations more clearly because the external field is insufficient to completely align the magnetic moments.

The χ^{-1} -T curves measured at 500 Oe and 50 Oe demonstrate that the magnetic response is field dependent. At 500 Oe, the stronger applied field suppresses spin disorder and promotes moment alignment, producing a more linear Curie-Weiss-like behavior. At 50 Oe, magnetic anisotropy and spin frustration become more evident, resulting in larger deviations from linearity. The positive Curie-Weiss temperature inferred from both curves confirms dominant ferrimagnetic exchange interactions. The broad transition region indicates the coexistence of ordered ferrimagnetic domains and thermally fluctuating spins, characteristic of nanocrystalline ferrites.

For $\text{Mg}_{0.2}\text{Mn}_{0.6}\text{Co}_{0.2}\text{Fe}_2\text{O}_4$, the increased Mn^{2+} concentration enhances the magnetic moment and modifies the cation distribution between A and B sites. The strong A–B superexchange interaction ($\text{Fe}^{3+}\text{--O}^{2-}\text{--Fe}^{3+}/\text{Mn}^{2+}$) promotes ferrimagnetic ordering, while surface spin disorder and finite particle-size effects lead to deviations from ideal Curie–Weiss behavior. Consequently, the $\chi^{-1}\text{--}T$ plots reveal a ferrimagnetic-to-paramagnetic transition accompanied by significant short-range magnetic correlations and magnetic anisotropy effects.

Figure 14 shows the temperature dependence of inverse magnetic susceptibility (χ^{-1}) for $\text{Mg}_{0.2}\text{Mn}_{0.6}\text{Co}_{0.2}\text{Fe}_2\text{O}_4$ measured under applied magnetic fields of 500 Oe and 50 Oe. In both cases, χ^{-1} increases with temperature and exhibits Curie–Weiss-like behavior in the high-temperature region, confirming the existence of a paramagnetic state above the magnetic ordering temperature. The positive Curie–Weiss temperature indicates predominant ferrimagnetic exchange interactions within the spinel lattice. Deviations from linearity at lower temperatures suggest the presence of short-range magnetic correlations, spin canting, and magnetic anisotropy effects. The broader curvature observed under 50 Oe compared to 500 Oe reflects enhanced sensitivity to spin disorder and magnetic fluctuations at low applied fields. These results confirm the ferrimagnetic nature of the $x = 0.6$ sample and highlight the significant role of Mn substitution in modifying the magnetic exchange interactions and thermal evolution of magnetization.

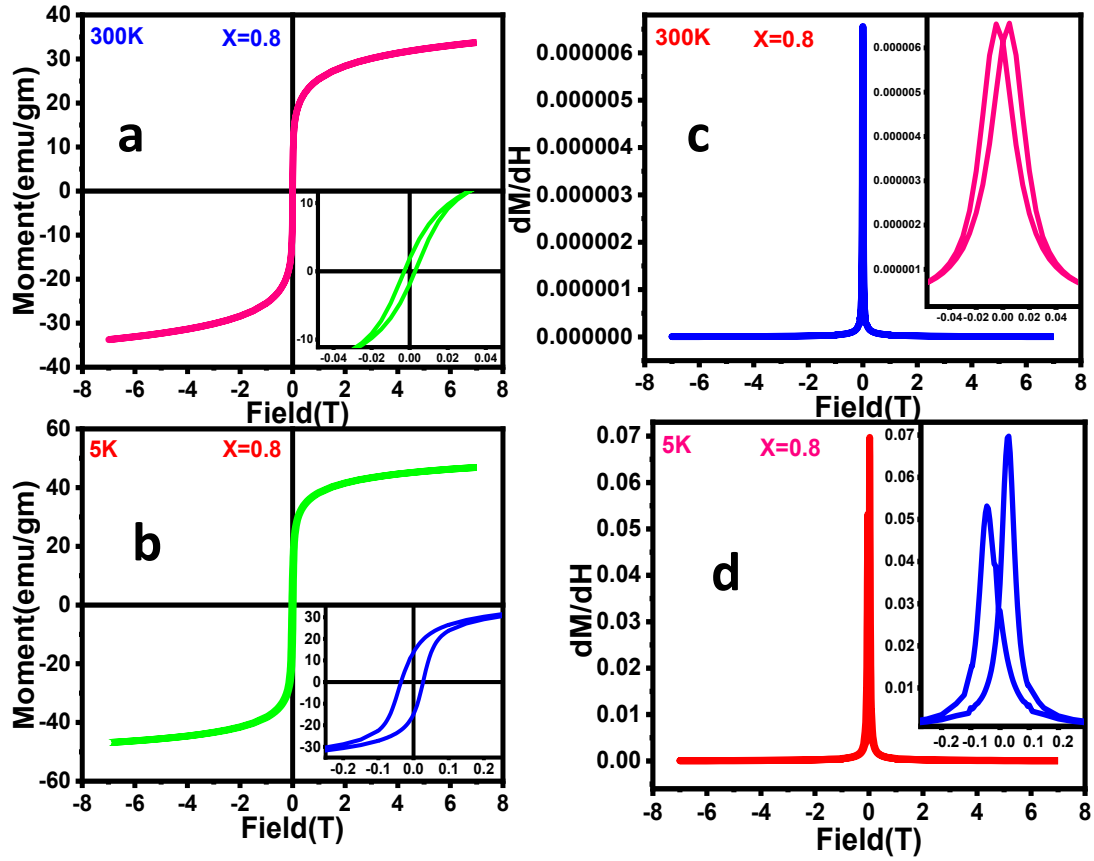


Figure:15. Magnetic hysteresis loops of $\text{Mg}_{0.2}\text{Mn}_{0.8}\text{Fe}_2\text{O}_4$ ferrite measured at (a) 300 K and (b) 5 K, along with the corresponding differential susceptibility curves (dM/dH) at (c) 300 K and (d) 5 K.

Figure 15 shows the magnetic hysteresis behavior of the $\text{Mg}_{0.2}\text{Mn}_{0.8}\text{Fe}_2\text{O}_4$ ferrite measured at 300 K and 5 K, together with the corresponding differential susceptibility curves (dM/dH). The results provide insight into the temperature-dependent magnetic ordering, anisotropy, and magnetization reversal mechanisms.

The hysteresis loop measured at 300 K exhibits a narrow S-shaped ferrimagnetic curve with rapid magnetization growth in the low-field region followed by gradual saturation at higher magnetic fields. The small loop opening indicates low coercivity and remanent magnetization, suggesting that thermal energy at room temperature facilitates spin rotation and domain-wall motion. The approach toward saturation confirms the presence of long-range ferrimagnetic ordering arising from strong A–B superexchange interactions between Fe^{3+} and Mn^{2+} ions. The

relatively soft magnetic behavior indicates reduced magnetocrystalline anisotropy and easier magnetization reversal. Compared with lower Mn concentrations, the $x = 0.8$ sample generally exhibits enhanced magnetic moment due to the larger magnetic moment of Mn^{2+} ions participating in the B-sublattice interactions.

At 5 K, the hysteresis loop becomes significantly wider than that observed at 300 K. The increase in coercivity and remanence indicates strengthening of magnetic anisotropy as thermal fluctuations are suppressed. The magnetization rises sharply at low fields and approaches saturation at high fields, confirming a well-established ferrimagnetic ground state. The higher saturation magnetization observed at 5 K suggests improved spin alignment and suppression of thermal spin disorder.

The differential susceptibility curve exhibits a single sharp peak centered near zero magnetic field. This behavior indicates that the largest change in magnetization occurs around the coercive field, where magnetic domains reverse most rapidly.

The derivative curve measured at 5 K displays two well-resolved symmetric peaks located on either side of zero field. These peaks correspond to the coercive fields during magnetization reversal. The increased peak separation compared with the room-temperature data indicates a substantial increase in coercivity. The symmetric nature of the peaks confirms stable ferrimagnetic ordering without evidence of exchange-bias effects.

The $M-H$ and dM/dH measurements demonstrate that the $x = 0.8$ composition exhibits robust ferrimagnetic behavior over the entire temperature range. The derivative curves further confirm that magnetization reversal occurs primarily through domain-wall movement and spin rotation. The transition from a narrow single dM/dH peak at 300 K to two separated peaks at 5 K reflects the development of stronger anisotropy and magnetic hardness at low temperature.

Figure 15 presents the M–H hysteresis loops and corresponding differential susceptibility curves for $\text{Mg}_{0.2}\text{Mn}_{0.8}\text{Fe}_2\text{O}_4$ ($x = 0.8$) measured at 300 K and 5 K. The room-temperature hysteresis loop exhibits soft ferrimagnetic behavior characterized by low coercivity and rapid magnetization saturation. At 5 K, the hysteresis loop becomes considerably wider, indicating enhanced coercivity, remanence, and magnetic anisotropy due to suppression of thermal fluctuations. The dM/dH curve at 300 K shows a narrow peak centered near zero field, confirming facile magnetization reversal and low anisotropy barriers. In contrast, the 5 K derivative curve displays two well-defined symmetric peaks separated by a larger field interval, reflecting increased coercive field and stronger domain-wall pinning. These observations demonstrate the strengthening of ferrimagnetic ordering and magnetic anisotropy with decreasing temperature in the Mn-rich $\text{Mg}_{0.2}\text{Mn}_{0.8}\text{Fe}_2\text{O}_4$ ferrite.

Figure 16 illustrates the temperature-dependent magnetization behavior of the $\text{Mg}_{0.2}\text{Mn}_{0.8}\text{Fe}_2\text{O}_4$ ($x = 0.8$) ferrite measured under applied magnetic fields of 500 Oe and 50 Oe using the zero-field-cooled (ZFC) and field-cooled (FC) protocols. The corresponding thermal irreversibility, represented by the difference between the FC and ZFC magnetizations (MFC–MZFC), is also presented. These measurements provide valuable information regarding the magnetic ordering, spin dynamics, blocking phenomena, and magnetic anisotropy of the Mn-rich ferrite sample.

As shown in Figure 16(a), the ZFC and FC magnetization curves measured under an applied field of 500 Oe exhibit a pronounced bifurcation over the entire temperature range investigated. The FC magnetization gradually decreases with increasing temperature, which is characteristic of a ferrimagnetically ordered system undergoing progressive thermal disordering. In contrast, the ZFC magnetization initially increases with temperature and reaches a broad maximum before decreasing at higher temperatures. The presence of a broad maximum in the ZFC curve

is indicative of a blocking process associated with nanocrystalline magnetic particles. The broad nature of the peak suggests a wide distribution of particle sizes, anisotropy energies, and magnetic relaxation times. The large separation between the FC and ZFC curves confirms significant magnetic irreversibility, reflecting strong magnetocrystalline anisotropy, domain-wall pinning, and spin disorder effects. The persistence of the FC–ZFC bifurcation up to room temperature indicates that the blocking temperature is higher than the measurement range, confirming the existence of a stable ferrimagnetic state.

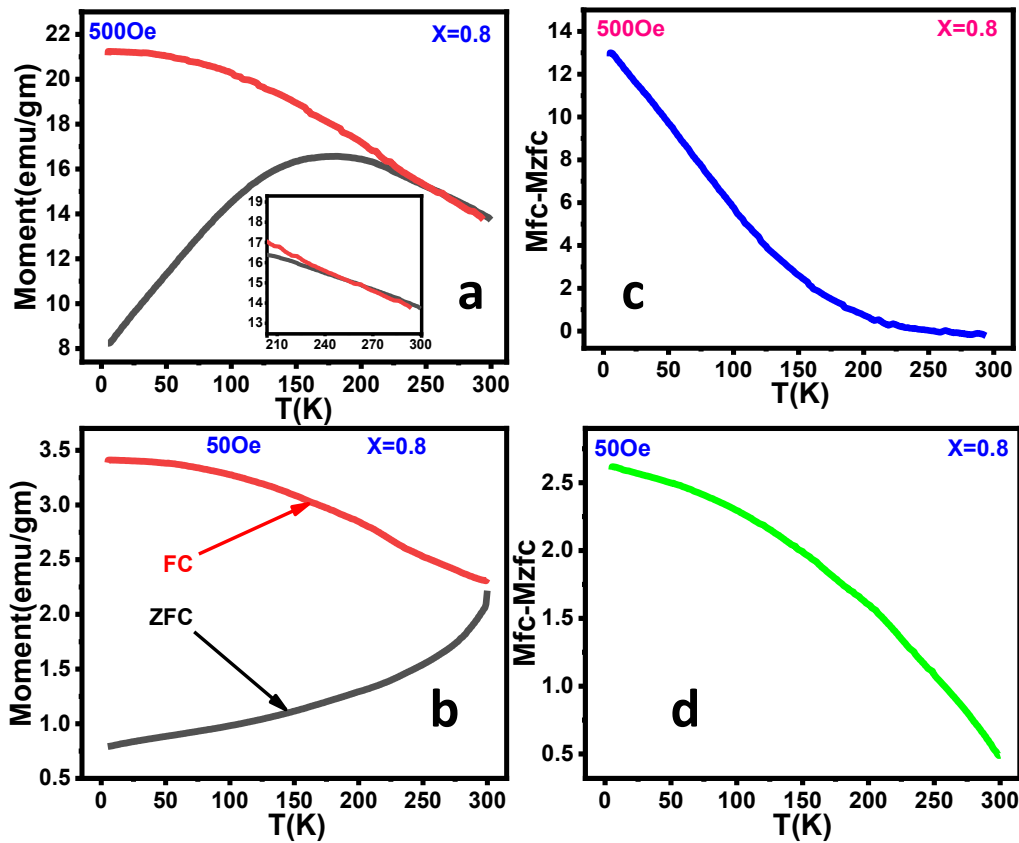


Figure:16. ZFC and FC magnetization curves of $\text{Mg}_{0.2}\text{Mn}_{0.8}\text{Fe}_2\text{O}_4$ ferrite measured under applied magnetic fields of (a) 500 Oe and (b) 50 Oe. The corresponding thermomagnetic irreversibility, expressed as $\Delta M = M_{FC} - M_{ZFC}$, is shown in (c) for 500 Oe and (d) for 50 Oe.

The corresponding thermal irreversibility, shown in Figure 16(c), further supports this interpretation. The difference between FC and ZFC magnetization initially increases with temperature, reaches a broad maximum in the intermediate temperature region, and

subsequently decreases at higher temperatures. The maximum in the MFC–MZFC curve represents the temperature region where the competition between thermal activation and magnetic anisotropy is strongest. At lower temperatures, the magnetic moments are frozen in metastable states, while increasing temperature enables partial relaxation of blocked spins. The gradual decrease in irreversibility at higher temperatures indicates enhanced thermal activation and reduced anisotropy barriers.

Figures 16(b) and 16(d) display the corresponding results obtained under a lower applied field of 50 Oe. The ZFC and FC curves exhibit an even larger separation compared with the 500 Oe measurements, demonstrating that magnetic irreversibility is significantly enhanced at lower fields. The reduced measuring field is insufficient to overcome anisotropy barriers and align the magnetic moments effectively, leading to stronger spin freezing and a more pronounced blocked state. The FC magnetization decreases monotonically with increasing temperature, whereas the ZFC curve remains considerably lower throughout the temperature range, confirming the existence of strong magnetic frustration and slow spin relaxation processes.

The MFC–MZFC difference curve measured at 50 Oe, shown in Figure 16(d), exhibits a continuously decreasing trend with increasing temperature. The large magnitude of the irreversibility at low temperatures indicates substantial spin freezing and strong anisotropy effects. As temperature increases, thermal fluctuations progressively assist spin relaxation and reduce the difference between FC and ZFC magnetizations. Nevertheless, the persistence of a finite irreversibility up to high temperatures confirms the presence of long-range ferrimagnetic interactions and significant magnetic anisotropy within the system.

Overall, the ZFC–FC measurements reveal that the $x = 0.8$ composition exhibits the strongest magnetic irreversibility among the investigated samples, which can be attributed to

the increased concentration of Mn^{2+} ions and the resulting enhancement of A–B superexchange interactions. The broad blocking features, large FC–ZFC separation, and substantial thermal irreversibility indicate the coexistence of ferrimagnetic ordering, surface spin disorder, and anisotropy-driven spin freezing. These observations demonstrate that Mn substitution plays a crucial role in strengthening the magnetic interactions and stabilizing the ferrimagnetic state in $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites.

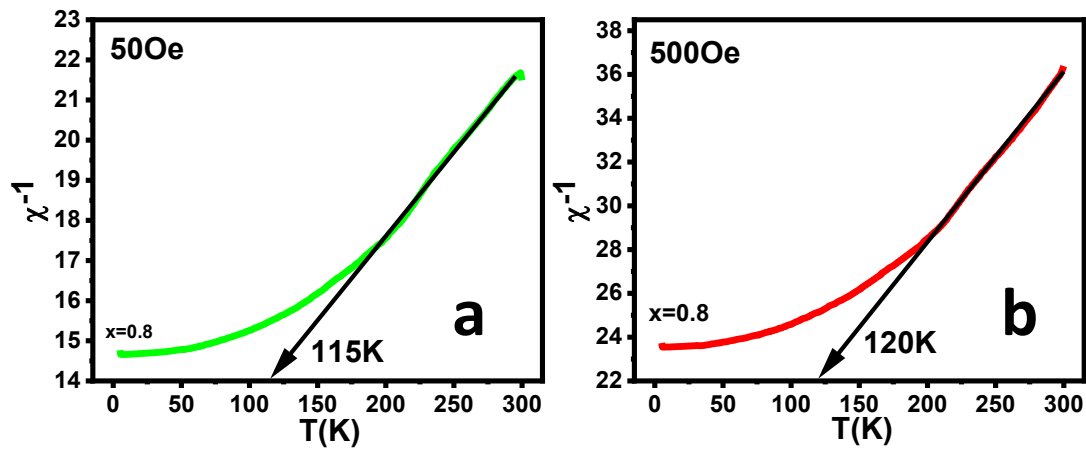


Figure:17. Temperature dependence of the inverse magnetic susceptibility (χ^{-1}) of $\text{Mg}_{0.2}\text{Mn}_{0.8}\text{Fe}_2\text{O}_4$ ferrite measured under applied magnetic fields of (a) 500 Oe and (b) 50 Oe.

Figure 17 presents the temperature dependence of the inverse magnetic susceptibility (χ^{-1}) for the $\text{Mg}_{0.2}\text{Mn}_{0.8}\text{Fe}_2\text{O}_4$ (x = 0.8) sample measured under applied magnetic fields of 500 Oe and 50 Oe. The χ^{-1} –T curves provide valuable information regarding the magnetic interactions and the nature of the magnetic phase transition in the ferrite system.

For the measurement performed under an applied field of 500 Oe (Figure 17a), the inverse susceptibility exhibits a gradual increase with increasing temperature over the entire investigated temperature range. At low temperatures, χ^{-1} remains relatively small due to the strong magnetic ordering and high susceptibility associated with the ferrimagnetic state. As the temperature increases, thermal fluctuations progressively weaken the exchange interactions

between the magnetic ions occupying the tetrahedral (A) and octahedral (B) sublattices, leading to a continuous reduction in susceptibility and a corresponding increase in χ^{-1} . The curve displays a nonlinear behavior over a broad temperature range and deviates from the ideal Curie–Weiss law expected for a simple paramagnetic system. Such deviation indicates the persistence of short-range magnetic correlations and spin-cluster interactions well above the magnetically ordered state. The upward curvature observed at higher temperatures suggests the gradual suppression of ferrimagnetic correlations rather than an abrupt magnetic transition. The extrapolated linear portion of the high-temperature region would yield a positive Weiss temperature, indicating the predominance of ferromagnetic or ferrimagnetic exchange interactions in the sample.

A similar trend is observed for the χ^{-1} –T curve measured under 50 Oe (Figure 17b), where the inverse susceptibility also increases continuously with temperature. However, compared with the 500 Oe measurement, the curve exhibits a more pronounced curvature throughout the temperature range. The stronger deviation from linear Curie–Weiss behavior under the lower applied field suggests that magnetic clusters, spin disorder, and local anisotropy effects have a greater influence when the external field is insufficient to fully align the magnetic moments. The low-field measurement is therefore more sensitive to intrinsic magnetic inhomogeneities and spin fluctuations present in the ferrite nanoparticles. The absence of a sharp discontinuity or divergence in χ^{-1} further confirms that the magnetic transition occurs over a broad temperature interval, which is characteristic of nanostructured ferrites possessing distributed anisotropy energies and finite-size effects.

The observed behavior of both χ^{-1} –T curves indicates that the $x = 0.8$ composition retains strong ferrimagnetic interactions throughout the investigated temperature range. The nonlinear temperature dependence and deviation from ideal Curie–Weiss behavior suggest the

coexistence of long-range ferrimagnetic ordering with short-range spin correlations, arising from competing superexchange interactions among Mn^{2+} , Co^{2+} , and Fe^{3+} ions. Furthermore, the enhanced curvature observed under the lower magnetic field highlights the significant role of spin canting, magnetic anisotropy, and thermally activated spin fluctuations in governing the magnetic response of the $x = 0.8$ ferrite sample. Overall, the χ^{-1} - T analysis confirms that Mn-rich $\text{Mg}_{0.2}\text{Mn}_{0.8}\text{Fe}_2\text{O}_4$ exhibits robust ferrimagnetic behavior accompanied by substantial magnetic correlations and spin-disorder effects, which become increasingly evident under low applied magnetic fields.

The Figure 18(a) shows the variation of dielectric loss tangent ($\tan \delta$) with frequency at 300 K. All compositions exhibit relatively high dielectric loss in the low-frequency region followed by a rapid decrease with increasing frequency, eventually approaching an almost constant value at higher frequencies. This behavior arises because charge carriers, mainly associated with electron hopping between $\text{Fe}^{2+}/\text{Fe}^{3+}$ ions and hole hopping between $\text{Co}^{2+}/\text{Co}^{3+}$ ions, can easily follow the slowly varying electric field at low frequencies, resulting in significant energy dissipation. As the frequency increases, the hopping charge carriers are unable to synchronize with the rapidly alternating field, leading to a reduction in polarization loss and consequently lower $\tan \delta$ values. Among the investigated samples, the $x = 0.4$ composition exhibits the highest dielectric loss, whereas the $x = 0.6$ sample shows the lowest loss over the entire frequency range, indicating improved dielectric stability and reduced energy dissipation.

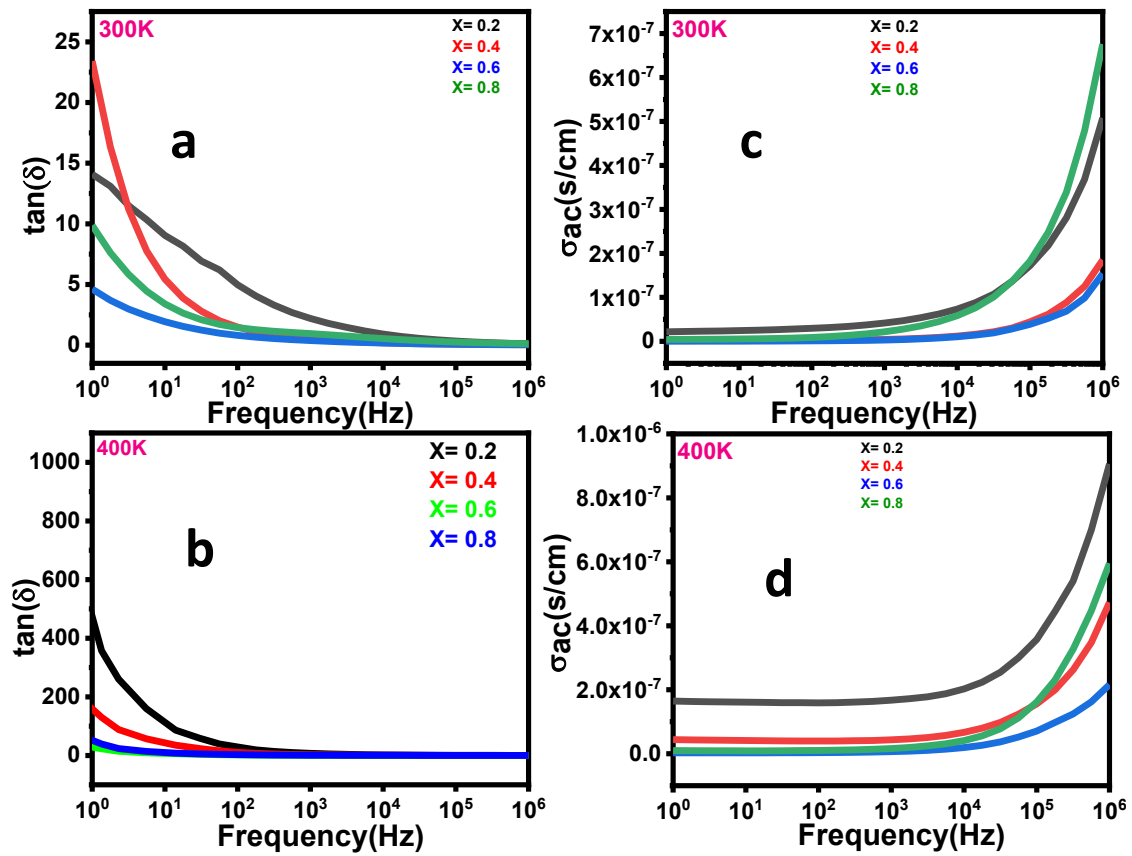


Figure:18. (a) The variation of dielectric loss tangent ($\tan \delta$) with frequency at 300 K. (b) The frequency dependence of $\tan \delta$ at 400 K is presented (c) The AC conductivity spectra measured at 300 K (d) The AC conductivity variation with frequency at 400 K.

The frequency dependence of $\tan \delta$ at 400 K is presented in Figure 18(b). A behavior similar to that observed at 300 K is maintained; however, the magnitude of dielectric loss increases noticeably due to enhanced thermal activation of charge carriers. The elevated temperature facilitates hopping conduction and interfacial polarization, thereby increasing dielectric relaxation losses. The $x = 0.8$ composition exhibits the highest $\tan \delta$ values at high frequencies, suggesting that increased Mn substitution enhances thermally activated hopping processes and space-charge polarization. The broad decrease of $\tan \delta$ with frequency further confirms the non-Debye nature of dielectric relaxation in these ferrite systems.

The AC conductivity spectra measured at 300 K are shown in Figure 18(c). The conductivity initially remains nearly constant at lower frequencies and then gradually increases

with increasing frequency. This behavior is characteristic of ferrite materials and can be explained by the hopping mechanism of localized charge carriers. At low frequencies, conductivity is dominated by grain boundaries, which act as resistive barriers to charge transport. With increasing frequency, the contribution from more conductive grains becomes dominant, leading to an increase in conductivity. The $x = 0.4$ composition exhibits the highest conductivity at 300 K, while the $x = 0.8$ sample shows comparatively lower conductivity, indicating that Mn substitution modifies the concentration and mobility of hopping charge carriers.

Figure 18(d) presents the AC conductivity variation with frequency at 400 K. The conductivity values are significantly higher than those observed at 300 K for all compositions, confirming the semiconducting nature of the ferrites. The enhancement in conductivity with temperature originates from thermally activated electron hopping between Fe^{2+} and Fe^{3+} ions located at octahedral sites. At higher frequencies, a sharp increase in conductivity is observed, consistent with Jonscher's universal power law behavior. The $x = 0.6$ composition exhibits the highest conductivity throughout most of the frequency range at 400 K, indicating an optimal balance between carrier concentration and hopping probability. The strong frequency and temperature dependence of conductivity demonstrates that charge transport in these ferrites is governed predominantly by thermally activated hopping conduction rather than band conduction.

Overall, the dielectric and conductivity results reveal that increasing frequency suppresses dielectric loss while enhancing AC conductivity. The temperature-induced increase in both $\tan \delta$ and conductivity confirms the semiconducting character of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites. The observed behavior is attributed to Maxwell–Wagner interfacial polarization, grain boundary effects, and thermally activated electron hopping between mixed-valence ions.

Furthermore, Mn substitution significantly influences the dielectric relaxation and charge transport mechanisms, making these ferrites promising candidates for high-frequency electronic and electromagnetic applications.

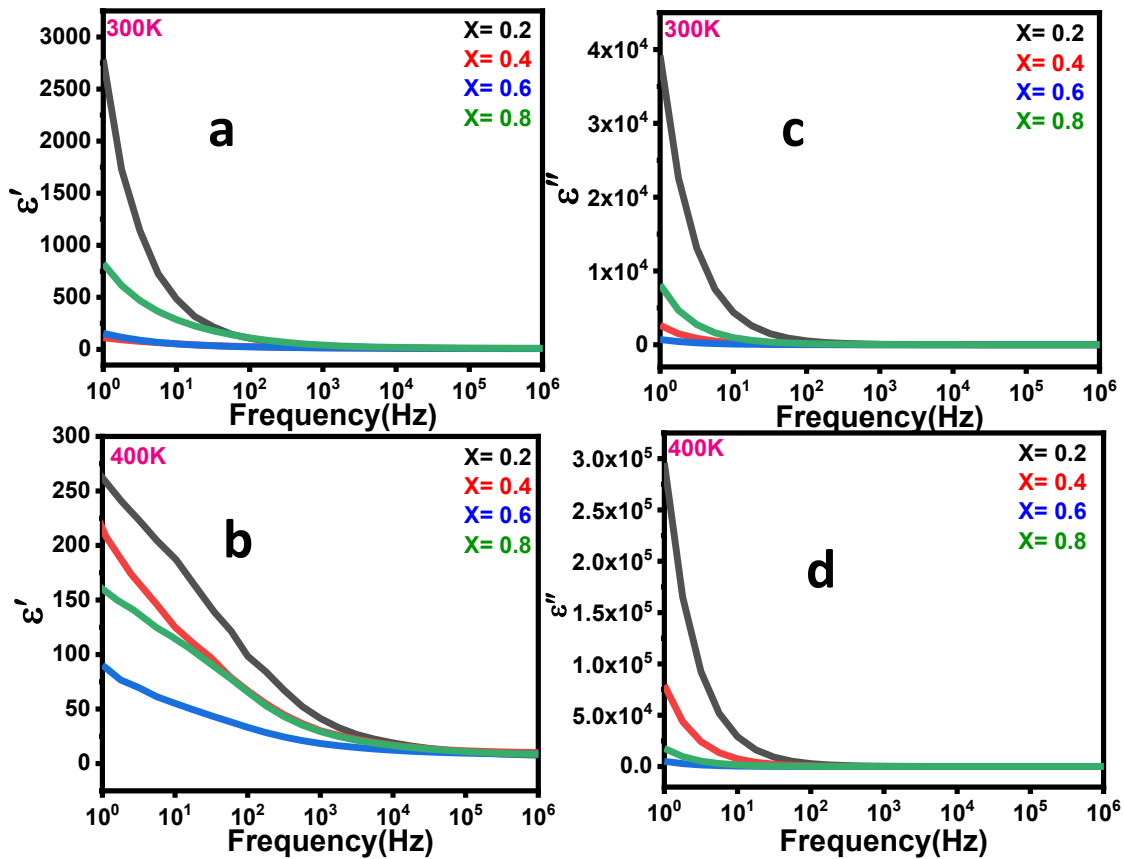


Figure:19. (a) The variation of the real part of dielectric permittivity (ϵ') as a function of frequency at 300 K. (b) The frequency dependence of ϵ' at 400 K. (c) illustrates the variation of the imaginary part of dielectric permittivity (ϵ'') with frequency at 300 K. (d) The imaginary dielectric permittivity measured at 400 K.

Figure 19 depicts the frequency dependence of the real (ϵ') and imaginary (ϵ'') parts of the dielectric permittivity for $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ($x = 0.4, 0.6, \text{ and } 0.8$) ferrites measured at 300 K and 400 K. The dielectric behavior is strongly influenced by interfacial polarization, charge carrier hopping, grain boundary effects, and temperature-dependent relaxation processes. The observed dielectric response can be satisfactorily explained using the Maxwell–Wagner polarization model together with Koop's two-layer theory, which considers ferrites as conducting grains separated by highly resistive grain boundaries.

Figure 19(a) presents the variation of the real part of dielectric permittivity (ϵ') as a function of frequency at 300 K. All compositions exhibit a high dielectric constant in the low-frequency region followed by a rapid decrease with increasing frequency, eventually reaching an almost frequency-independent value at higher frequencies. This behavior is characteristic of ferrite materials and arises from the strong contribution of space-charge polarization at low frequencies. In this region, charge carriers accumulate at grain boundaries and interfaces, resulting in large polarization and consequently high dielectric constant values. As the frequency increases, the charge carriers become unable to follow the rapidly alternating electric field, leading to the suppression of interfacial polarization and a decrease in ϵ' . Among the investigated samples, the $x = 0.4$ composition exhibits the highest dielectric constant throughout the frequency range, whereas the $x = 0.6$ composition shows the lowest dielectric response, indicating a reduced contribution of space-charge polarization.

The frequency dependence of ϵ' at 400 K is shown in Figure 19(b). The overall behavior remains similar to that observed at 300 K, with ϵ' decreasing continuously with increasing frequency. However, the magnitude of the dielectric constant is generally enhanced due to the increased mobility of charge carriers at elevated temperature. Thermal energy promotes electron hopping between Fe^{2+} and Fe^{3+} ions and facilitates polarization processes, particularly at low frequencies. The enhancement of dielectric constant at higher temperatures confirms the thermally activated nature of polarization in these ferrites. The persistence of frequency dispersion at 400 K further indicates the existence of dielectric relaxation associated with charge carrier dynamics and grain boundary effects.

Figure 19(c) illustrates the variation of the imaginary part of dielectric permittivity (ϵ'') with frequency at 300 K. The dielectric loss component exhibits large values at low frequencies and decreases rapidly with increasing frequency for all compositions. The high ϵ'' values

observed in the low-frequency region originate from conduction losses associated with charge carrier hopping and space-charge accumulation at grain boundaries. As the frequency increases, these mechanisms become less effective because the hopping electrons cannot respond to the rapidly changing electric field, resulting in a significant reduction in dielectric loss. The $x = 0.4$ sample exhibits the highest ϵ'' values, indicating stronger polarization and greater energy dissipation, whereas the $x = 0.6$ composition shows comparatively lower dielectric loss.

The imaginary dielectric permittivity measured at 400 K is presented in Figure 19(d). Compared with the corresponding 300 K data, the ϵ'' values are significantly enhanced, particularly in the low-frequency region. This increase arises from thermally activated hopping conduction and the increased participation of charge carriers in polarization processes. The pronounced low-frequency dispersion confirms the dominant role of grain boundary polarization and charge accumulation. As the frequency increases, ϵ'' decreases sharply and approaches a nearly constant value, indicating that dielectric relaxation mechanisms become progressively ineffective at higher frequencies. The $x = 0.4$ composition again exhibits the largest dielectric loss, while the $x = 0.6$ sample maintains the lowest loss over the measured frequency range.

Overall, the dielectric spectra reveal a typical dispersion behavior in both ϵ' and ϵ'' , characterized by high values at low frequencies and a gradual decrease with increasing frequency. This behavior confirms the dominance of Maxwell–Wagner interfacial polarization and Koop's grain boundary model in governing the dielectric response of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites. The increase in dielectric parameters with temperature indicates thermally activated polarization and hopping conduction mechanisms. Furthermore, Mn substitution significantly influences the dielectric response by modifying charge carrier

concentration, grain boundary resistance, and electron hopping probability between mixed-valence ions. The combined dielectric results suggest that these ferrites possess strong frequency-dependent polarization characteristics, making them suitable for applications in high-frequency electronic devices, electromagnetic absorbers, sensors, and energy-storage systems.

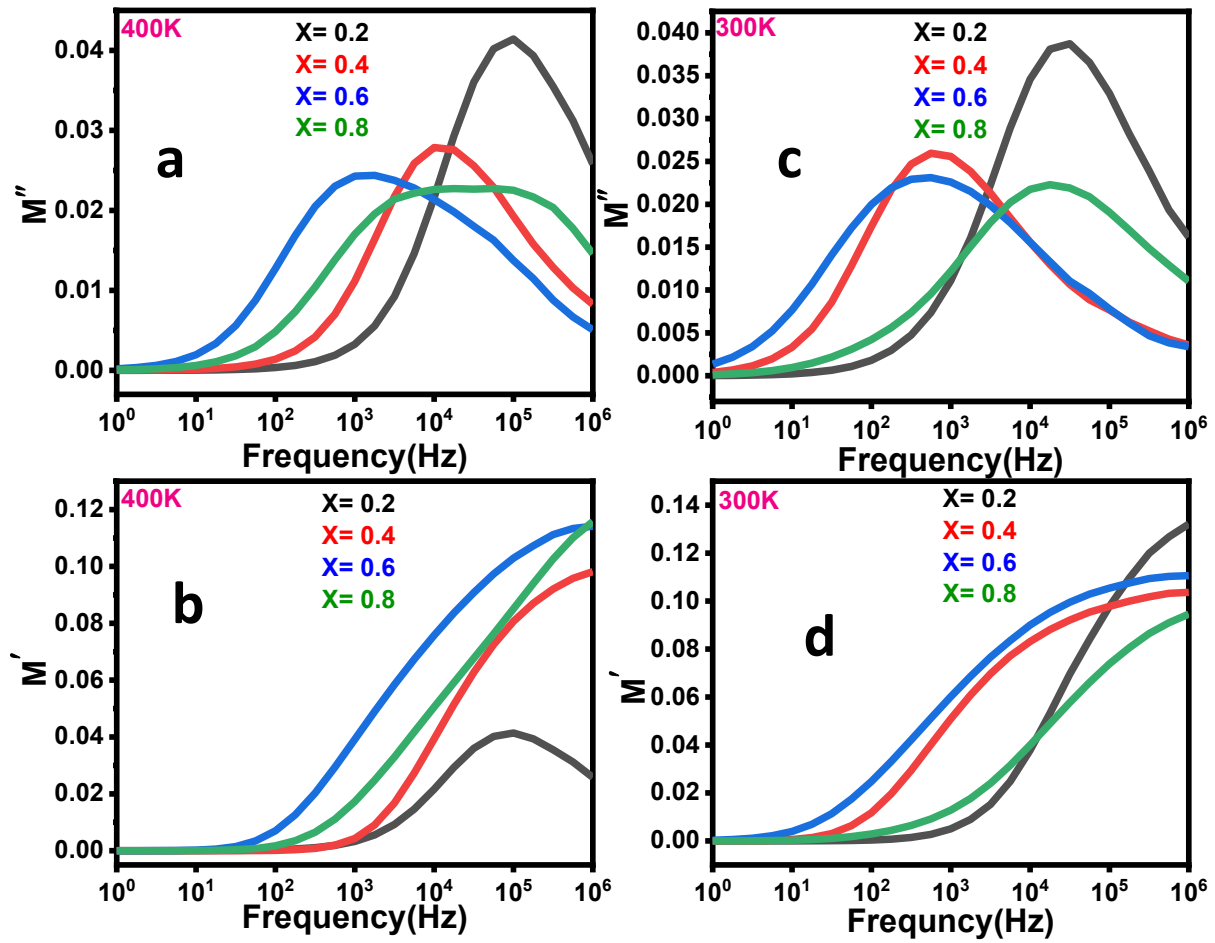


Figure:20. (a) The variation of the imaginary part of electric modulus (M'') with frequency at 400 K. (b) The variation of the real part of electric modulus (M') with frequency at 400 K (c)The real part of the electric modulus (M') at 300 K.(d) The variation of M' with frequency at 300 K.

Figure 20 illustrates the frequency-dependent electric modulus spectra of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites ($x = 0.2, 0.4, 0.6$, and 0.8) measured at 300 K and 400 K. The electric modulus formalism is particularly useful for suppressing the effects of electrode polarization and emphasizing the relaxation behavior associated with charge carrier dynamics

within the grains. The real modulus (M') and imaginary modulus (M'') spectra provide valuable information regarding charge transport, dielectric relaxation, and hopping mechanisms in the investigated ferrites.

Figure 20(a) presents the variation of the imaginary part of electric modulus (M'') with frequency at 400 K. All compositions exhibit broad asymmetric relaxation peaks, indicating the presence of non-Debye type relaxation arising from a distribution of relaxation times rather than a single relaxation process. The broadness of the peaks suggests that multiple charge carriers and localized hopping mechanisms contribute simultaneously to the dielectric relaxation. The relaxation peak position varies with Mn concentration, indicating that the relaxation time is strongly influenced by cation distribution and charge carrier mobility. The $x = 0.2$ composition exhibits the highest M'' peak intensity and the peak appears at relatively higher frequency, indicating a shorter relaxation time and faster charge carrier dynamics. With increasing Mn substitution, the peak position gradually shifts and becomes broader, suggesting increased disorder and enhanced interaction among localized charge carriers. The shift of the relaxation peak toward lower frequencies for some compositions signifies longer relaxation times due to stronger localization of hopping electrons.

The corresponding real part of the electric modulus (M') at 300 K is shown in Figure 20(c). At low frequencies, M' remains close to zero for all samples, indicating negligible restoring force against charge carrier motion and confirming that long-range charge transport dominates in this region. As the frequency increases, M' increases continuously and approaches a saturation value at higher frequencies. This behavior reflects the transition from long-range hopping conduction at low frequencies to localized or short-range mobility at higher frequencies. The increase in M' suggests that charge carriers gradually lose their ability to follow the rapidly oscillating electric field. Among the investigated samples, variations in the

growth rate and saturation values indicate differences in relaxation dynamics caused by Mn-induced modifications in the $\text{Fe}^{2+}/\text{Fe}^{3+}$ hopping concentration and microstructural characteristics.

Figure 20(b) shows the M'' spectra measured at 400 K. Compared with the 300 K results, the relaxation peaks become more pronounced and generally shift toward higher frequencies. This shift is characteristic of thermally activated relaxation processes, where increasing temperature enhances charge carrier mobility and reduces relaxation time. The movement of the M'' maxima toward higher frequencies confirms that the hopping of electrons between Fe^{2+} and Fe^{3+} ions becomes easier at elevated temperatures. Furthermore, the increased peak intensity at 400 K indicates enhanced dielectric relaxation resulting from increased thermal activation of charge carriers. The persistence of broad relaxation peaks even at higher temperature confirms that the relaxation mechanism remains non-Debye in nature.

The variation of M' with frequency at 400 K is presented in Figure 20(d). Similar to the behavior observed at 300 K, M' remains very small at low frequencies and increases significantly with increasing frequency. However, the rise in M' occurs at comparatively lower frequencies than at room temperature, reflecting the enhanced conductivity and reduced relaxation time caused by thermal activation. The higher modulus values observed at elevated temperature indicate increased participation of charge carriers in polarization processes. The temperature-induced enhancement of M' further supports the existence of thermally activated hopping conduction mechanisms within the ferrite lattice.

Overall, the electric modulus analysis confirms that dielectric relaxation in $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites is governed by thermally activated hopping of charge carriers, predominantly through $\text{Fe}^{2+} \leftrightarrow \text{Fe}^{3+}$ electron exchange at octahedral sites. The broad relaxation peaks and frequency-dependent modulus behavior reveal a non-Debye relaxation process with

a distribution of relaxation times. Increasing temperature facilitates charge carrier mobility, leading to shorter relaxation times and enhanced dielectric response. The observed composition-dependent variations demonstrate that Mn substitution significantly influences the charge transport and relaxation dynamics by modifying cation distribution, hopping probability, and localized polarization within the spinel ferrite structure.

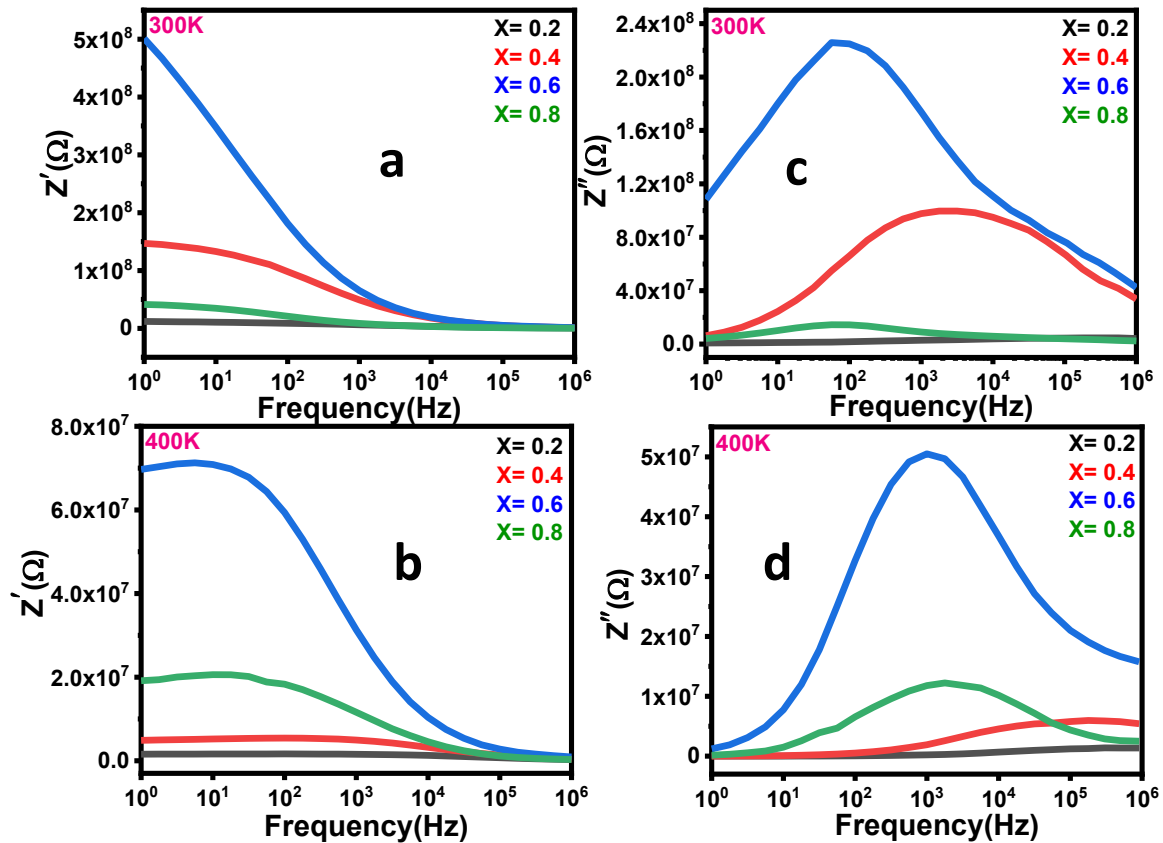


Figure:21. (a) The frequency dependence of the real part of impedance (Z') at 300 K (b) The frequency dependence of Z' at 400 K (c) The variation of the imaginary part of impedance (Z'') with frequency at 300 K (d) The variation of Z'' with frequency at 400 K.

Figure 21 illustrates the complex impedance behavior of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites ($x = 0.2, 0.4, 0.6, \text{ and } 0.8$) measured at 300 K and 400 K. The frequency dependence of the real and imaginary components of impedance provides valuable information regarding charge transport, grain boundary effects, and dielectric relaxation processes occurring within the ferrite system.

The frequency dependence of the real part of impedance (Z') at 300 K is presented in Figure 21(a). A broad relaxation peak is observed for the $x = 0.4$ and $x = 0.6$ compositions, whereas the $x = 0.2$ and $x = 0.8$ samples exhibit much weaker responses. The appearance of these broad maxima indicates dielectric relaxation associated with electron hopping between Fe^{2+} and Fe^{3+} ions located at octahedral sites. The broad nature of the peaks suggests a non-Debye type relaxation process characterized by a distribution of relaxation times rather than a single relaxation mechanism. The highest Z' peak intensity observed for the $x = 0.6$ composition reflects enhanced localization of charge carriers and stronger polarization effects. The shift and variation of peak intensities with composition indicate that Mn substitution significantly modifies the electrical microstructure and relaxation dynamics of the ferrite system.

The variation of the imaginary part of impedance (Z'') with frequency at 300 K is shown in Figure 21(c). All samples exhibit high Z'' values in the low-frequency region followed by a continuous decrease with increasing frequency, eventually approaching very small values at higher frequencies. This behavior is characteristic of ferrite materials and indicates the presence of space-charge polarization and grain boundary resistance at low frequencies. The large impedance observed at lower frequencies arises from the accumulation of charge carriers at grain boundaries, which act as highly resistive regions. As the frequency increases, the hopping electrons between Fe^{2+} and Fe^{3+} ions become unable to follow the rapidly alternating electric field, resulting in a significant reduction in Z'' . Among the investigated compositions, the $x = 0.6$ sample exhibits the highest Z'' values, suggesting a larger grain-boundary contribution and higher resistive character compared with the other compositions. The $x = 0.2$ sample shows the lowest impedance values over the entire frequency range, indicating relatively easier charge transport.

The frequency dependence of Z' at 400 K is shown in Figure 21(b). Well-defined relaxation peaks become more prominent at elevated temperature, particularly for the $x = 0.6$ composition, while weaker peaks are observed for $x = 0.4$ and $x = 0.8$. The increase in peak intensity and the enhanced relaxation behavior at higher temperatures indicate that thermally activated charge carriers contribute more effectively to polarization processes. The broad relaxation maxima further confirm the non-Debye nature of the electrical relaxation. The presence of temperature-dependent peaks suggests that the relaxation mechanism is governed by hopping conduction and interfacial polarization effects. The dominant response observed for the $x = 0.6$ sample implies an optimum balance between charge carrier concentration and grain-boundary resistance, resulting in stronger dielectric relaxation.

Figure 21(d) displays the variation of Z'' with frequency at 400 K. Compared with the behavior at 300 K, the impedance values decrease substantially for all samples, demonstrating enhanced electrical conductivity at elevated temperatures. The reduction in impedance is attributed to thermally activated hopping of charge carriers between $\text{Fe}^{2+}/\text{Fe}^{3+}$ and $\text{Co}^{2+}/\text{Co}^{3+}$ ions. Increased thermal energy facilitates carrier mobility and lowers the effective grain-boundary resistance. The $x = 0.6$ composition continues to exhibit the highest impedance values, although the overall magnitude is considerably reduced. The faster decrease of Z'' at higher temperatures confirms the semiconducting nature of the ferrites and indicates negative temperature coefficient of resistance (NTCR) behavior.

Overall, the impedance spectra reveal that the electrical transport in $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites is strongly frequency and temperature dependent. The decrease in both Z' and Z'' with increasing frequency and temperature confirms semiconducting behavior governed by thermally activated hopping conduction. The broad relaxation peaks observed in the impedance spectra indicate non-Debye relaxation associated with heterogeneous microstructures

consisting of conducting grains separated by resistive grain boundaries. Among the studied compositions, the $x = 0.6$ sample exhibits the most pronounced impedance and relaxation features, suggesting enhanced grain-boundary polarization and stronger charge carrier localization compared with the other compositions. These results are consistent with the Maxwell–Wagner interfacial polarization model and Koop's phenomenological theory, which successfully explain the dielectric and electrical behavior of spinel ferrite materials.

3.4. Fourier Transform Infrared (FTIR) spectroscopy

The Fourier Transform Infrared (FTIR) spectra of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites ($x = 0.0, 0.2, 0.4, 0.6, \text{ and } 0.8$) recorded in the range of $400\text{--}4000\text{ cm}^{-1}$ are shown in **Figure 22**. The spectra exhibit the characteristic absorption bands of spinel ferrites, confirming the successful formation of the ferrite phase. Two prominent absorption bands are observed in the low-wavenumber region, which are associated with the intrinsic vibrations of metal–oxygen bonds at the tetrahedral (A) and octahedral (B) sites of the spinel lattice.

The strong absorption band appearing around $550\text{--}600\text{ cm}^{-1}$ is attributed to the stretching vibrations of metal–oxygen (M–O) bonds at the tetrahedral sites (ν_1), while the band located near $400\text{--}450\text{ cm}^{-1}$ corresponds to the octahedral-site vibrations (ν_2). The presence of these characteristic bands confirms the formation of the cubic spinel structure in all compositions. Generally, the tetrahedral vibration band appears at a higher frequency than the octahedral band because the bond length at the tetrahedral site is shorter, resulting in a larger force constant.

With increasing Mn concentration, slight shifts in the absorption bands can be observed. These shifts are associated with the replacement of Co^{2+} ions by Mn^{2+} ions, leading to changes in cation distribution, bond lengths, and force constants within the spinel lattice. Since Mn^{2+} possesses a larger ionic radius than Co^{2+} , substitution induces local lattice distortions and

modifies the metal–oxygen bonding environment. The gradual variation in band position therefore provides evidence for the successful incorporation of Mn ions into the ferrite matrix.

In addition to the ferrite-related bands, weak absorption features observed around **3400–3600 cm^{-1}** are attributed to O–H stretching vibrations originating from adsorbed moisture and hydroxyl groups on the particle surface. The band near **1600–1650 cm^{-1}** corresponds to the bending vibration of molecular water. A weak feature around **2300–2400 cm^{-1}** may be assigned to atmospheric CO_2 absorbed during spectral acquisition. The relatively low intensity of these bands indicates the effective removal of residual organic constituents during calcination.

The increase in intensity and sharpening of the ferrite bands with Mn substitution suggest improved crystallinity and enhanced ordering of the spinel structure. The absence of significant impurity-related absorption bands further confirms the phase purity of the synthesized samples. Overall, the FTIR results corroborate the XRD findings and verify the successful synthesis of single-phase $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ nanocrystalline ferrites.

The observed FTIR spectra reveal the characteristic tetrahedral and octahedral vibrational modes of spinel ferrites, confirming the formation of the $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ phase. The slight shifts in the metal–oxygen stretching bands with increasing Mn content indicate modifications in the cation distribution and local bonding environment caused by Mn^{2+} substitution. These changes are consistent with the larger ionic radius of Mn^{2+} compared to Co^{2+} and suggest a gradual alteration of the super-exchange pathways responsible for the magnetic behavior of the ferrites. The absence of impurity-related absorption bands demonstrates the successful synthesis of phase-pure nanocrystalline ferrites through the sol–gel auto-combustion method.

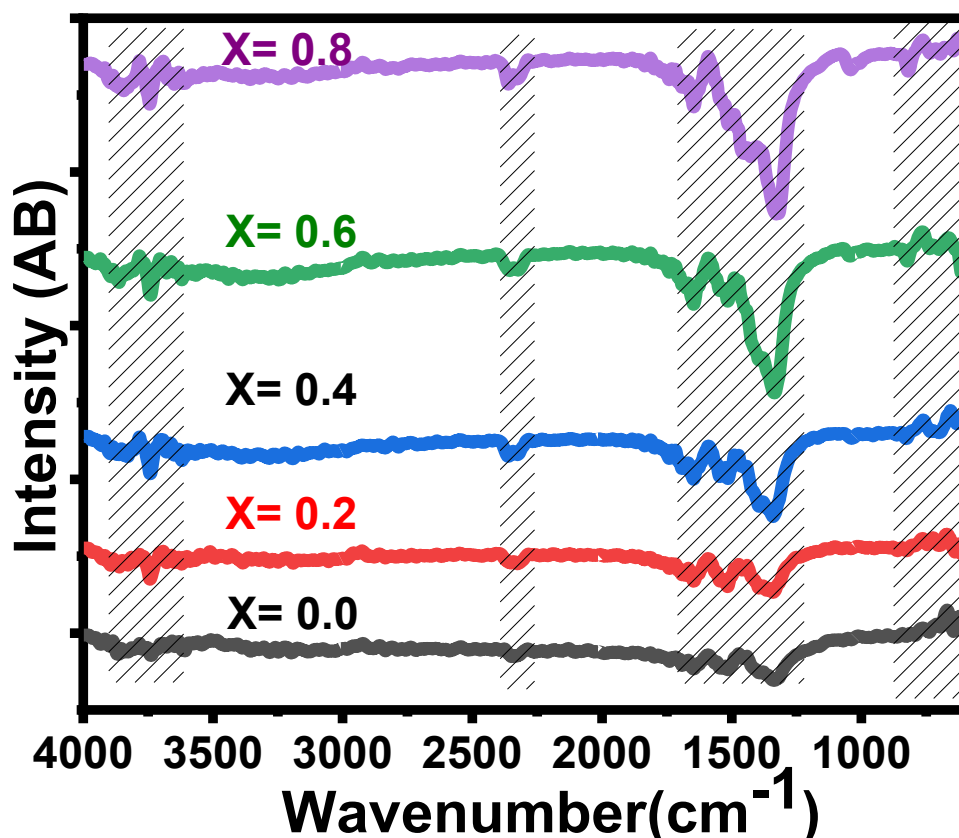


Figure:22. The Fourier Transform Infrared (FTIR) spectra of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites ($x = 0.0, 0.2, 0.4, 0.6, \text{ and } 0.8$) recorded in the range of $400\text{--}4000\text{ cm}^{-1}$.

3.5. Raman Spectroscopy

Raman spectroscopy shows the atomic structure and chemical bonds in the (where $x = 0.2, 0.4, 0.6, 0.8$) ferrite samples. A 532 nm laser beam was used for the analysis of the Raman spectrum of the Ni substituted ferrites. The Raman spectra reveal stretching vibrations of spinel ferrites within the $300 - 690\text{ cm}^{-1}$ range, as shown in Figure 23. The mode observed at 323 cm^{-1} is ascribed to metal ions vibration in octahedral (B) coordination within the spinel lattice. While the higher-frequency mode at 684 cm^{-1} is related with M-O stretching in tetrahedral (A-site) coordination. The intermediate bonds at 457 cm^{-1} are associated with the mixed vibrational contribution of both octahedral (B-site) and tetrahedral (A- site). When multiple cationic species contribute to a Raman mode (as exemplified in M-O_4), the resultant spectral peak exhibit broadening and incorporates contributions from various cations possessing identical symmetry. Consequently, through meticulous deconvolution of the spectral peaks, the integrated intensity beneath each subpeak functions as an indicator of the concentration of each cation within the unit cell.

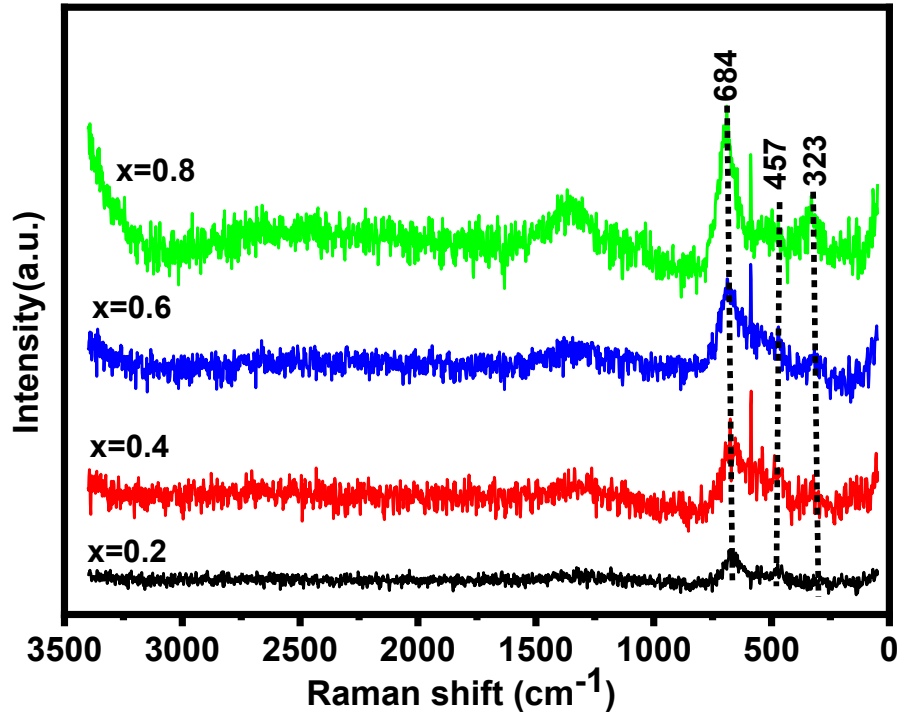


Figure:23. Raman Spectra of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites ($x = 0.0, 0.2, 0.4, 0.6, \text{ and } 0.8$)

There are two possible configurations for the metallic cations: either encircled by four oxygen ions or encircled by six oxygen ions, making an octahedron. Typically, for a material to show Raman active, its nuclear displacement-induced variation in polarizability, denoted as da/dq , must be non-zero. Three prominent peaks consistently appear in the Raman spectra of the synthesized spinel ferrites within the $200\text{--}800\text{ cm}^{-1}$ range. In figure 24, A_{1g} near 700 cm^{-1} represents the metal oxygen bond of symmetric stretching vibration of tetrahedral A-site. T_{2g} (3) around 500 cm^{-1} represents asymmetric vibrational stretching involving octahedral (B-site) metal oxygen bonds. E_g near 300 cm^{-1} shows symmetric bending vibrations of oxygen atom in spinel lattice.

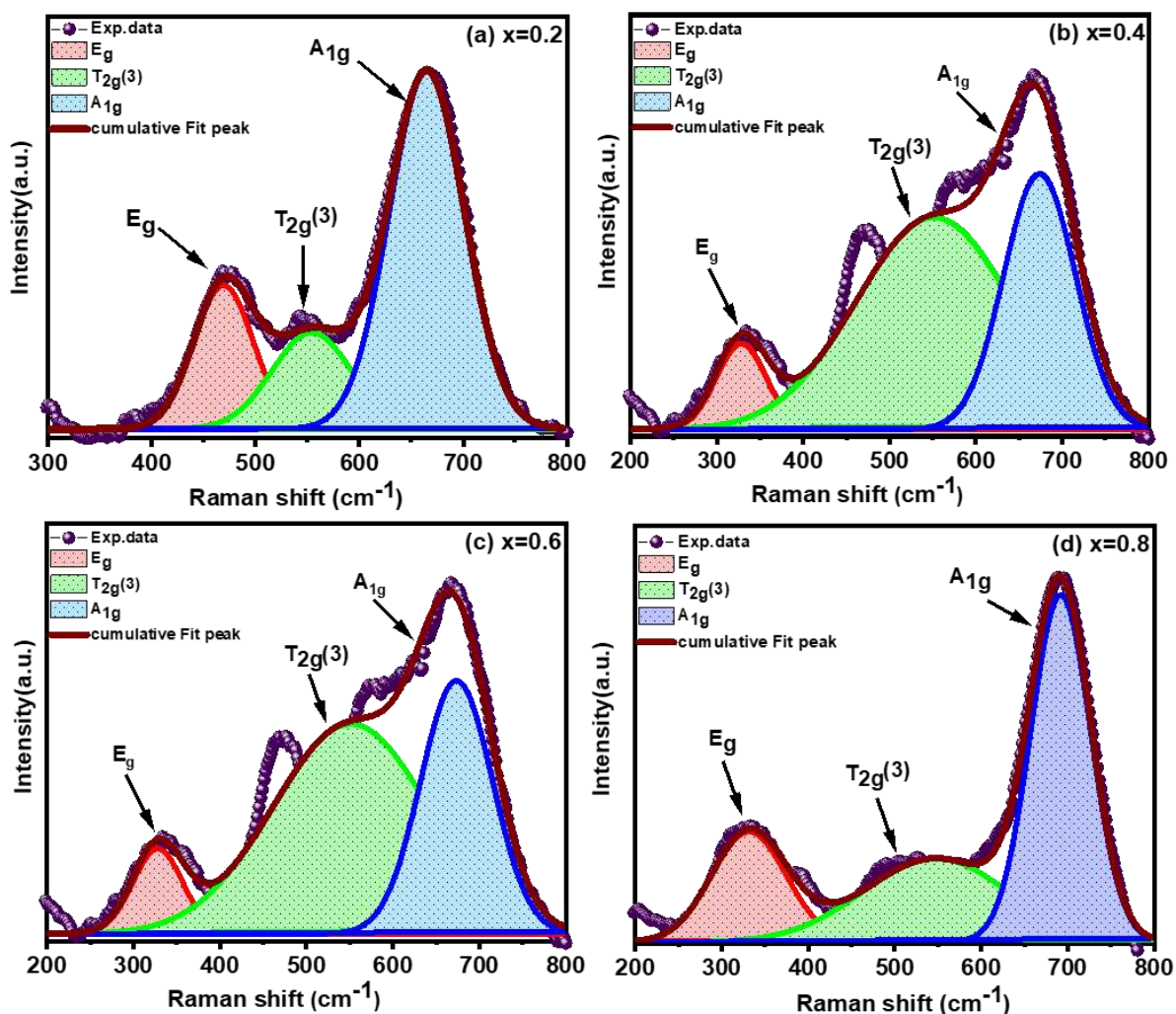


Figure: Deconvoluted Raman Spectra of $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites ($x = 0.0, 0.2, 0.4, 0.6,$ and 0.8).

4. Conclusions

Nanocrystalline $\text{Mg}_{0.2}\text{Mn}_x\text{Co}_{0.8-x}\text{Fe}_2\text{O}_4$ ferrites ($x = 0.0\text{--}0.8$) were successfully synthesized by the sol–gel auto-combustion technique. XRD and Rietveld refinement analyses confirmed the formation of a phase-pure cubic spinel structure for all compositions. The crystallite size decreased from about 50 nm to 20 nm with increasing Mn content, while the lattice parameter and unit-cell volume increased at higher Mn concentrations due to the substitution of larger Mn^{2+} ions for Co^{2+} ions. The calculated hopping lengths at tetrahedral and octahedral sites increased systematically with Mn incorporation, indicating modification of the interionic distances and magnetic exchange pathways. Williamson–Hall analysis revealed the

presence of tensile lattice strain, whereas the reduction in dislocation density suggested improved crystallinity. FESEM and AFM investigations confirmed the nanocrystalline nature of the ferrites and revealed composition-dependent variations in particle agglomeration, grain morphology, and surface roughness. FTIR spectroscopy further verified the formation of the spinel ferrite phase through the observation of characteristic metal–oxygen stretching vibrations associated with tetrahedral and octahedral sites. Magnetic measurements demonstrated ferrimagnetic behavior throughout the investigated composition range. M–H loops recorded at 300 K exhibited soft magnetic characteristics with low coercivity, while measurements at 5 K revealed enhanced coercivity, remanence, and magnetic anisotropy due to suppression of thermal fluctuations. The dM/dH analysis indicated a transition from domain-wall-motion-dominated magnetization reversal at room temperature to anisotropy-controlled pinning mechanisms at low temperature. ZFC–FC measurements showed strong thermomagnetic irreversibility, significant interparticle interactions, and anisotropy-driven spin freezing, with the irreversibility increasing at lower applied fields. Overall, Mn substitution effectively modifies the cation distribution, lattice structure, and magnetic exchange interactions, providing a viable route for tailoring the magnetic performance of Mg–Co ferrite nanoparticles for technological applications in magnetic storage, sensors, microwave devices, and spintronic systems.

Data availability

Not Applicable.

Declarations

Authors do not have potential conflicts of interest. The authors have no relevant financial or non-financial interests to disclose.

Supplementary information: Not Applicable.

Ethical approval: Not applicable.

Data availability

Not Applicable.

CRedit authorship contribution statement

P. M. Saini: Sample preparation, methodology, writing original draft, **Shobha Ram Choudhary:** investigation, methodology, **Ritu:** Conceptualization, experimental work, **N.S. Leel:** methodology, investigation, **Yogesh Kumar Yadav:** methodology, experimental work, **Rajan Mishra:** methodology, experimental work, **Saurabh Dalela:** investigation, methodology, **P. A. Alvi:** Investigation, data analysis, **B. L. Choudhary:** writing original draft, conceptualization, Review & editing and supervision.

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